October 1953

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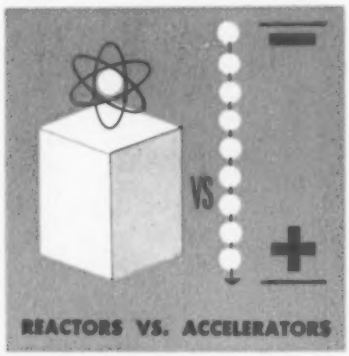

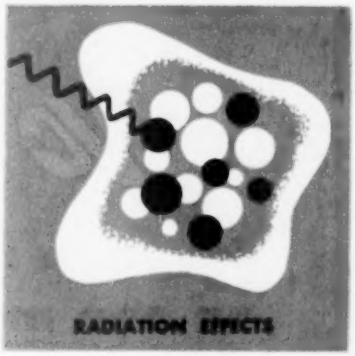

RADIATION STERILIZATION

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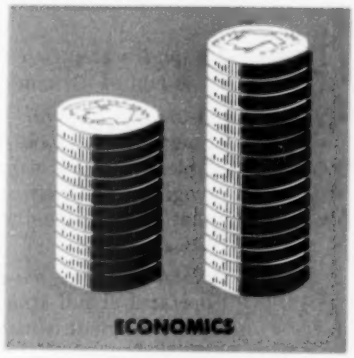
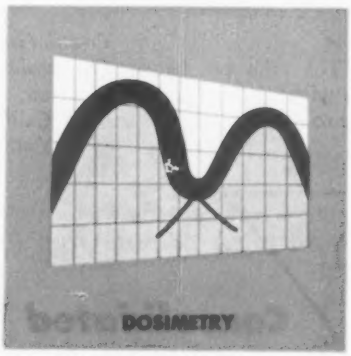
THE PROBLEMS WE FACE





See page 7 for first articles in an extensive series on Radiation Sterilization

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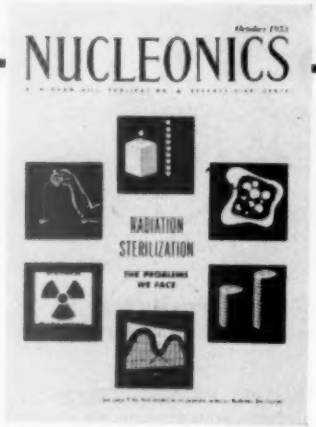

OCTOBER • 1953

Vol. 11, No. 10

Radiation Sterilization: Introduction... 7

Fission-Product Potential of Commercial Reactors and Their Processes... 8
DICK DUFFEY

Van de Graaff Accelerators for Sterilization Use... 14
F. L. FOSTER, D. R. DEWEY II, and A. J. GALE

The Industrial Future of Radiation Chemistry... 18
BERNARD MANOWITZ

Boron Detection in Tissues Using the (n,α) Reaction... 21
GIOVANNA MAYR, H. D. BRUNER, and MARSHALL BRUCER

Problems in Operating a Research Reactor... 26
ROBERT W. POWELL

Birmingham Nuclear Physics Conference... 32
JOHN K. BELING

Monitoring of Water for Fission-Product Contamination... 36
W. R. LOOSEMORE

Measurements of Gamma-Ray Dosage in Finite Absorbers... 41
ERNEST J. HENLEY

Radiation Decomposition of Carbon-14-Labeled Compounds... 44
RICHARD M. LEMMON

Fluorine 18: Preparation, Properties, Uses... 46
RICHARD B. BERNSTEIN and JOSEPH J. KATZ

Radiophotoluminescence Dosimetry System of the U. S. Navy... 52
JAMES H. SCHULMAN, WILLIAM SHURCLIFF, ROBERT J. GINTHER, and FRANK H. ATTIX

Cross Sections

Automatic Sample Changer and Recorder for Dipping Counters... 58
B. P. McKAY and D. B. ZILVERSMIT

Radioactive Contamination Sampling by Smears and Adhesive Disks... 60
EUGENE V. BARRY and LEONARD R. SOLON

Low-Cost I^{131} Thyroid Uptake Apparatus... 64
EDWARD SIEGEL

Separate Assay of Fe^{55} and Fe^{59} in Mixed Preparations... 66
W. B. STEWART and H. H. ROSSI

Fast and Simple γ-Ray Scintillation Spectrometer... 68
GERALD J. HINE

Departments

EDITORIAL... 7
BOOKS... 70
NEWS... 72
LETTERS... 78
PRODUCTS AND MATERIALS... 84

JEROME D. LUNTZ, Editor
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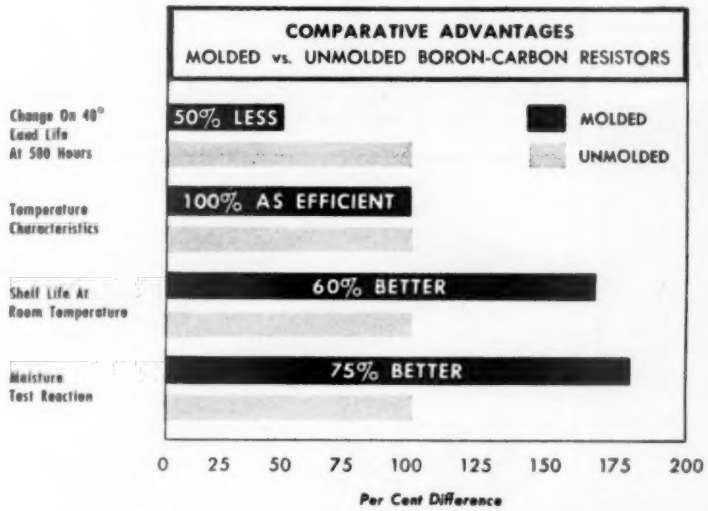

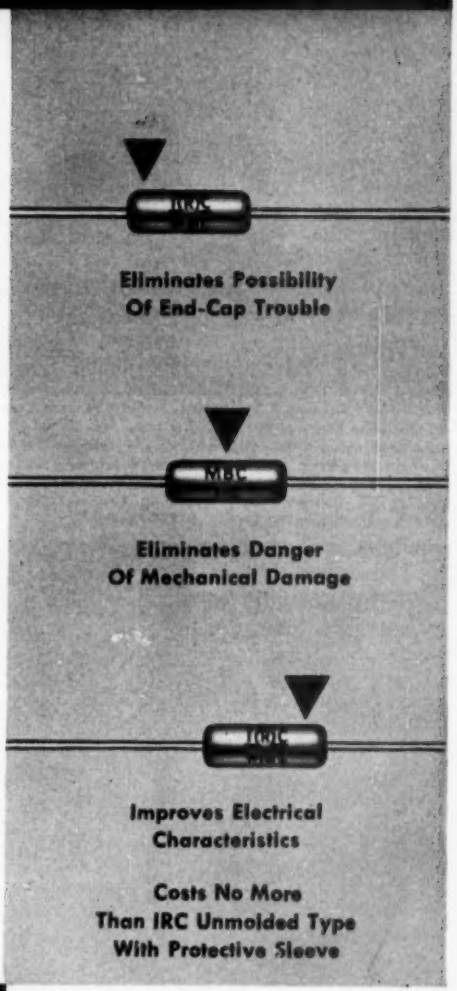
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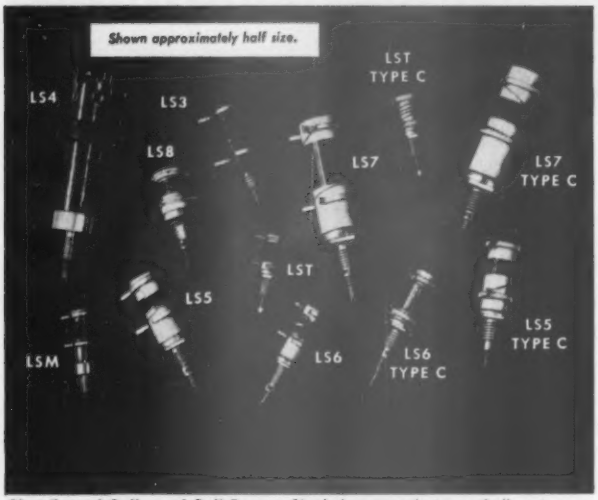

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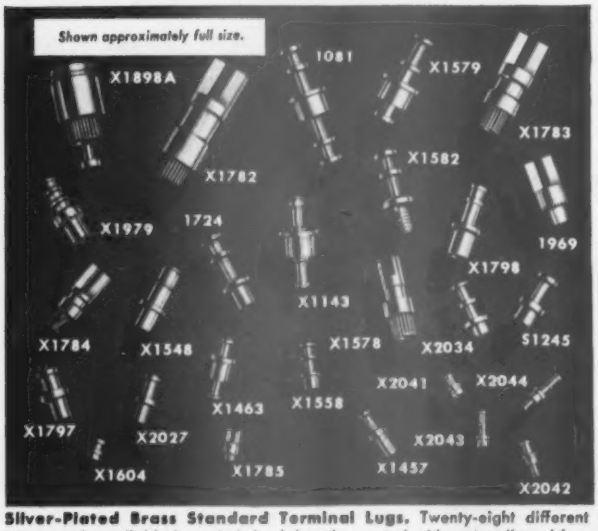

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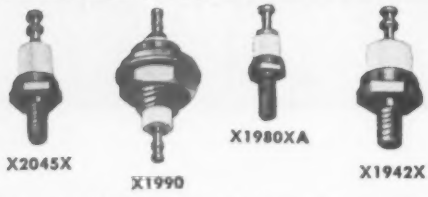

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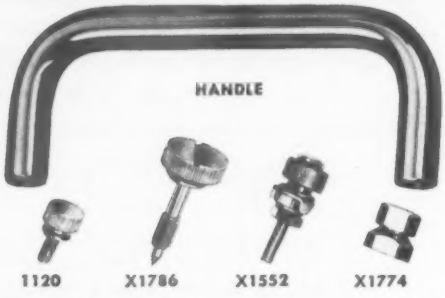

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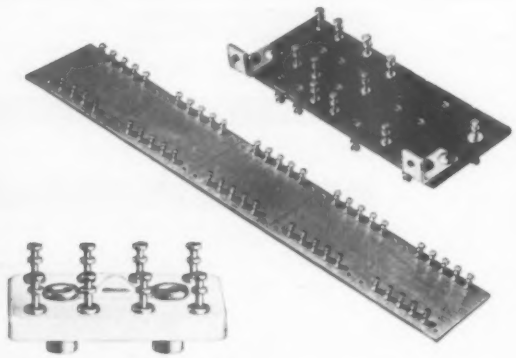

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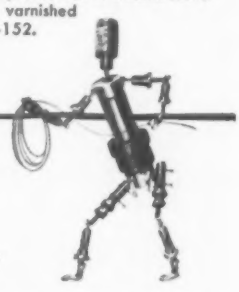

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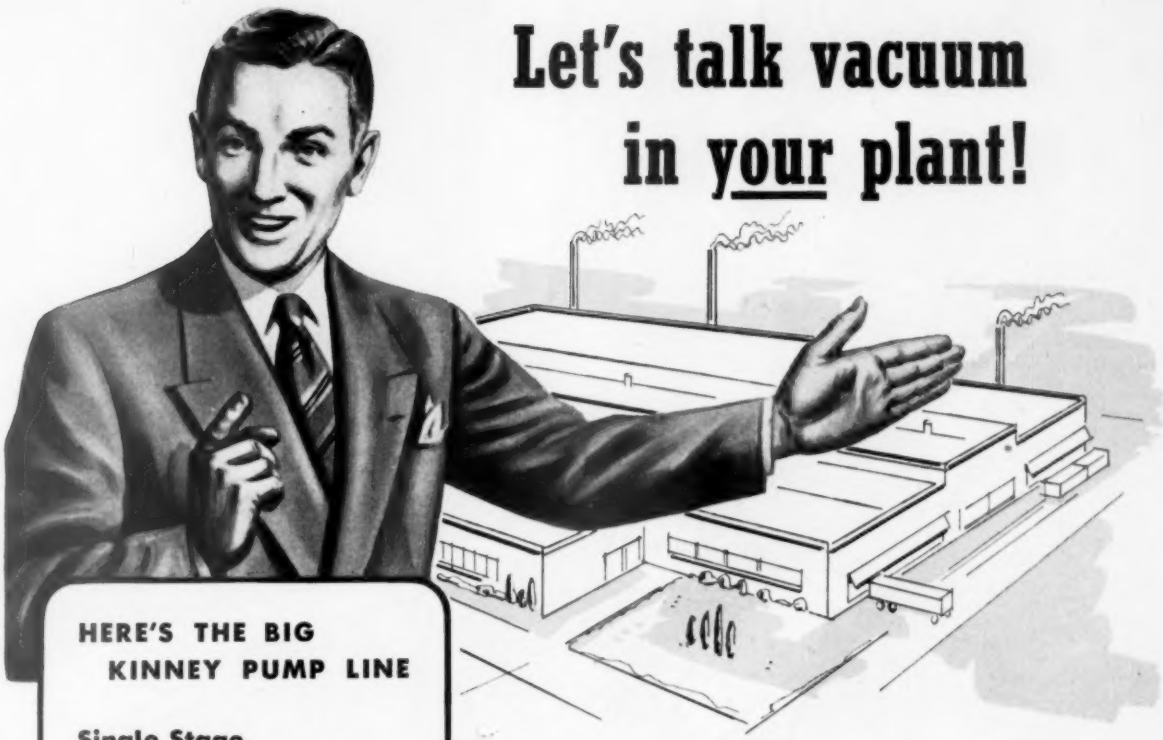

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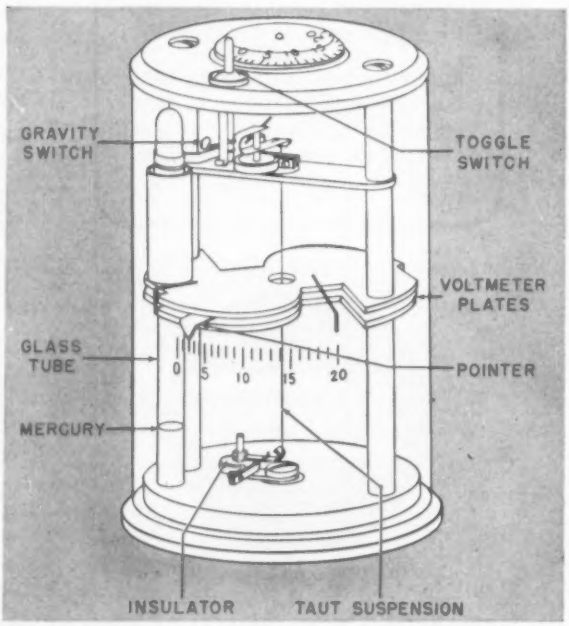
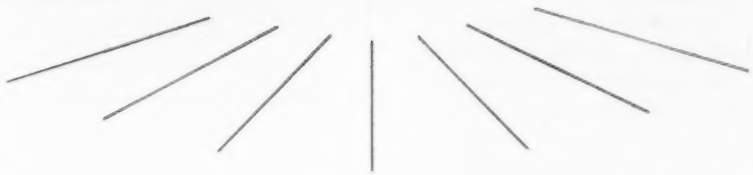


CUTAWAY VIEW of monitor shows operating parts. Self-charging gravity switch is operated by inverting the monitor.

Radiation Sterilization



SINCE SHORTLY AFTER the discovery of X-rays by Roentgen in 1895, scientists have attempted to utilize the bactericidal properties of ionizing radiations. However, it has only been in very recent years, with the development of particle accelerators and nuclear reactors, that the possibilities have become intriguing. Why is such a radically different means of sterilization of such tremendous interest, and where does this interest lie?

In the food industry, frozen foods have developed a wide degree of acceptability. Their chief drawbacks are the refrigeration requirements for storage and the attendant cost. In many military operations and installations, refrigeration is either unavailable or limited in capacity. Although the past 20 years have shown great improvements in the thermal processing of canned foods with attendant improvements in quality, there are still a number of canned food items today, such as certain canned meat products, that require a quantity of heat energy for sterilization that is far in excess of the amount that would result in optimal flavor and texture. In addition, the gage of metal required in the containers for heat sterilization could be greatly reduced for a heatless sterilization process.

Among the many heat-sensitive pharmaceuticals used in vast quantities today are the metabolic endproducts of microorganisms, such as the antibiotics. As they are heat-sensitive, they cannot be sterilized by thermal means, and since they are produced from microorganisms, the difficulties in obtaining sterile products are obvious. Accordingly, there is great hope that radiation sterilization will prove useful.

HOWEVER, there are a number of problems. For sterilization purposes, energy requirements appear to be of the order of 16.6 joules per gram or 1.66×10^8 ergs per gram. This represents an equivalent energy absorption of 1.05×10^{20} electron-volts per gram. At the rate of 25 ev as a requirement for breaking a C—H bond in, for example, a CH_2 group (containing 1.3×10^{22} bonds per gram), a dose of two million roentgens (16.6 joules/gram) could result in the breakage of 4×10^{18} bonds per gram or approximately 0.003% of the bonds present.

If approximately the same percentage of bonds are broken in organic components of foods when these are exposed to equivalent doses of ionizing energy, it is interesting to speculate on the results. First of all, changes should occur, although very small, percentage-wise. (That such changes do occur has been demonstrated.) Even our modern physical and chemical techniques are not, for the most part, sensitive enough to detect changes of such small magnitude as represented by 0.003%. However, the important point to consider is whether changes of such small quanta will have a physiological effect on those consuming the irradiated product.

Herein lies the great point of difference between foods and drugs insofar as radiation

sterilization is concerned. Some foods are consumed at least thrice daily, every day in the week, whereas drugs are consumed only infrequently and in relatively small quantities, by any one individual, and then usually under the order and control of a physician.

In addition to possible physiological implications, there are other problems: These deal with the sources of ionizing energy—accelerator vs. isotopic, beta vs. gamma radiations, reliability of production of the radiations, efficiency of utilization, personnel safety, shielding problems, etc. Insofar as the products are concerned, there are a number of factors that must be considered, evaluated and studied further—sterility of the products and the factors influencing it, taste and flavor, acceptability and a study of various means for improving the same, packaging problems, authorization by governmental regulatory bodies.

A major problem for industry is the economics of radiation sterilization. What will radiation sources cost? In what sizes will they be available? How would the development of a civilian nuclear power industry affect the economics of isotopic sources?

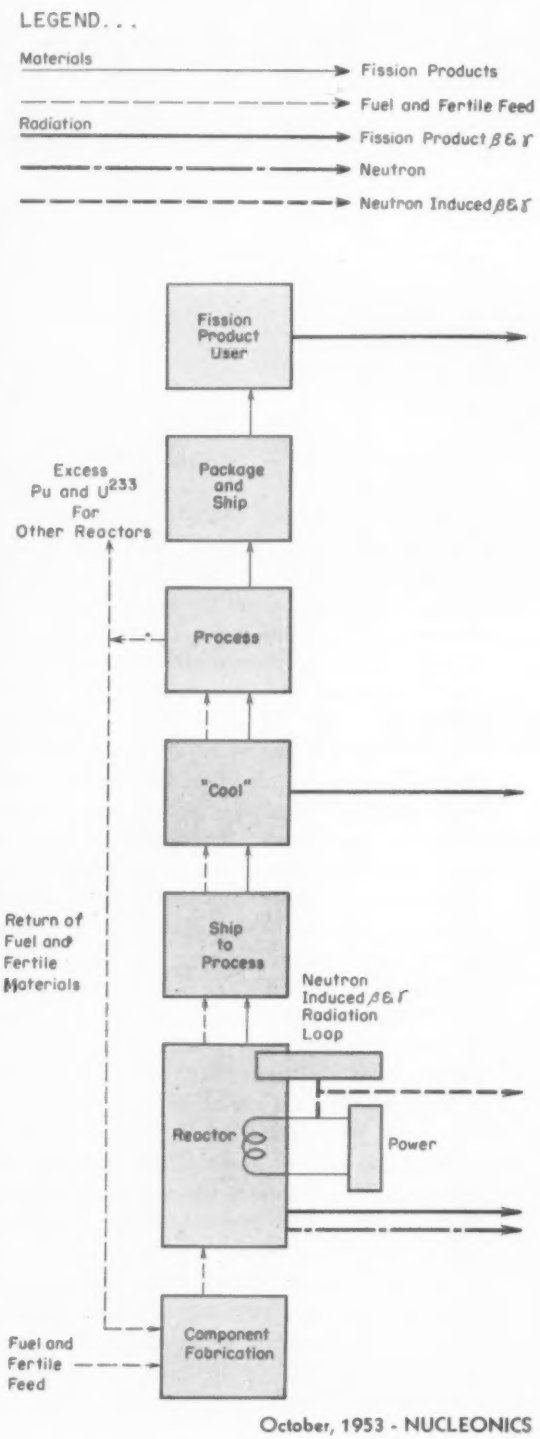
Undoubtedly the greatest spur to the resolution of the economic problem would be the development of important applications of radiation.

BECAUSE OF THE GREAT SIGNIFICANCE to our readers of this phase of the atomic energy field, the editors of NUCLEONICS have arranged with Prof. S. A. Goldblith, Dept. of Food Technology, Massachusetts Institute of Technology, to obtain an extensive series of articles. Written by people working in the field, the articles—about 12—will cover the technical, economic and legal aspects of radiation sterilization. We also intend to present the viewpoints of industry, the government and the military.

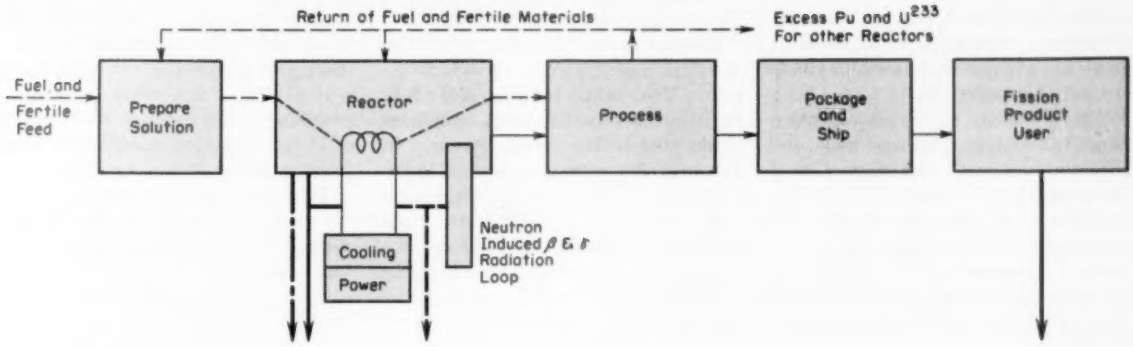
We hope that this series will serve as the first progress report on what may yet be the first significant peacetime byproduct of our atomic energy program.

THE COVER is a representation of six broad problems that must be solved before radiation sterilization can become an industrial process. Dosimetry is represented by a curve of ionization produced by double irradiation in an accelerator.

RADIATION STERILIZATION I



SOURCES OF RADIATION from projected commercial reactor complexes. At left is solid-system reactor with processing; below, solution fluid reactor with integral processing



Fission-Product Potential of Commercial Reactors and Their Processes

One of the primary potential sources of radiation for sterilization is the nuclear reactor and its fission products. On the basis of current knowledge, future reactor development possibilities are related to sterilization requirements

By DICK DUFFEY
Division of Reactor Development
U. S. Atomic Energy Commission
Washington, D. C.

SALE OF POWER is now the basis for considering commercial reactors; however, some feel that fission products, the co-product of reactor power, may ultimately be of significant value. But the situation with fission products is quite different from reactor power in that the competition is not defined. This results from the fact that there are no large established uses for beta and gamma radiation (apparently fission product's "useful" property) which, of course, has heretofore been unavailable in quantity. Therefore, the potentialities for fission products are quite speculative. It should be noted that there will undoubtedly be reactor income from neutron radiation, such as sale of artificial isotopes, but this will not be considered in this article.

Some interest in fission products may stem from certain aspects of the profit picture. The profits from reactor power, with the exception of that internally used by a manufacturing organization, may be controlled by a public regulatory group. On the other hand, the profits from fission products used indirectly in manufacturing products, such as chemicals or foods, may not be limited in this way at all.

Many commercial uses for fission products have been suggested. Those of potentially the greatest economic importance, like sterilization of foods, would require very large quantities of fission-product radiation. Although such large quantities can be foreseen when atomic energy contributes substantially to the country's power, the amounts of available fission-product radiation will be very much dependent on the type of reactors and, particularly, the type of separation processes used. This results from the fact that much of the fission-product radiation is from relatively short-lived materials and is lost from use by either long irradiation of fuels in the reactor or delayed processing and packaging.

To be sure that sufficient quantities, as well as satisfactory kinds, of fission-product sources are available, the possible users should, as soon as prac-

ticable, place a value on fission products. This would insure that commercial reactors and processes are developed, designed, and operated under the premise that fission products are valuable materials.

The following will briefly mention suggested fission product uses and potential demands and then discuss the amounts of fission products that might be available from reactors and processes. Any recommendation of a particular reactor or processing scheme to produce fission-product radiation must be based on future economic studies.

Amounts of Radiation Necessary

Relatively large amounts of fission-product radiation would be required for many of the proposed uses. Tables 1, 2, and 3 relate a few values of dosage, production of certain materials proposed for treatment, and estimated treatment rates, respectively. Experimenters emphasize the fact that much development must be done to establish fully the feasibility of proposed uses. The following figures are only to indicate source requirements should these larger scale uses develop. (Apparently some believe that more specialized applications, e.g., biological sterilizations, which would require much smaller quantities of fission products, may be practical at an earlier date than the larger-scale uses.)

To give some idea of the magnitude of fission-product requirements indicated, let us assume that 10% of the annual pork and milk production were sterilized at the Vitro rates (Table 3) and that the sources are in use a third of the time; then about 4 and 90 Mw gross-fission-product sources would be required, respectively. Enzyme deactivation would require more while only partial sterilization (maybe pasteurization) would require less. One might argue over the utilization efficiencies, which depend on the "shielding" power of the material treated, the geometry of the system, and the effective time that a source is in use. But this does not affect the conclusion that these larger-scale applications would require many megawatts, possibly hundreds, of fission-product power.

Reactor Sources of Radiation

Fission-product radiation can be obtained within the reactor core, from used fuel elements, from separated fission products, and from the exit fuel stream of fluid-fuel reactors. This is illustrated on pages 8 and 9.

Reactor core. The reactor itself is, of course, a very intense source of radiation. The total gamma and beta radiation at equilibrium is something under 10% of the reactor power. Of this over 6% is fission-product radiation, and roughly half of this is gamma (*11*). However, it is felt that the necessary shielding to reduce the neutron flux to avoid activating the materials being treated would reduce the available gammas and betas considerably (*12*). Even if sufficient gamma flux were made available, it seems likely that the reactor designer would find it difficult to provide for such radiations.

Such use of the reactor core will not be discussed further because the following sources appear more attractive at present to the author. No doubt this will receive more study in the future by reactor designers.

TABLE 1—Radiation Dosage for Suggested Applications

	Approximate dose (rep)*
Sterilization of foods	2,000,000 (*1*)
Enzyme deactivation in foods	10,000,000 (*1*)
Insect control in grain	25,000 (*2*)
Polymerization of styrene	10,000,000 (*3*)

* A rep, roentgen equivalent physical, is 93 ergs of radiation energy absorbed per gram.

TABLE 2—Examples of Production of Materials Suggested for Radiation Treatment

Material	*Approximate 1949 production (lb × 10^6)*
Canned fruits and vegetables	10,000 (*4*)
Dressed beef	10,000 (*5*)
Dressed pork	10,000 (*5*)
Whole milk	80,000 (*6*)
Fermented malt beverages	20,000 (*7*)
Wheat	60,000 (*8*)
Styrene	300 (*9*)

Used fuel. Discharged fuel (or fertile material like U^{238}) is a fission-product package of relatively high power since the decay is short, although still probably an average of several days, minimum. Handling problems would seem to limit this to solid fuel elements or solidified liquid fuels, rather than packaged liquids.

The fission-product power expected in used fuel from certain reactor designs is indicated later under "Future Reactor Complexes." In any case, it seems that fuels enriched in fissionable materials, i.e., U^{235}, Pu, and U^{233}, with their probably higher fission-product radioactivity would seem to be preferable.

Such used-fuel sources are mobile, but the location of central processing plants and the reactors result in shipping complications. The user has little control over the fission-product package, which may not be a very useful design for radiations. Processing "cooling" requirements (retention of material prior to processing to facilitate operations) would eliminate some fuel inventory charges to the user.

Separated fission products from used fuels. Most consideration has been given to packaged fission products from the reactor processing wastes. There will be losses in any source preparation, and, of course, production of large quantities is not established.

This scheme provides the most mobility and results in the prepared source having the least dependence on other operations of the reactor complex, i.e., fuel preparation, reactor operation, and processing. In general, with solid-system reactors, "cooling," processing (possibly in central plants), source preparation, and shipping times will probably result in a decay of months. Presumably, this might be shortened by new methods, the justification being reduced processing costs, particularly reduced inventory charges, and the value of the increased amounts of radiation. Fluid reactors with continuous processing would provide the freshest separated fission products of all schemes.

Exit fluid fuel stream. The external fuel circuit of a fluid reactor—used fuel in a way—provides fission products with essentially no decay, since pumping times are usually a matter of seconds. Normally, in an externally cooled fluid reactor, this radiation would be lost as heat.

TABLE 3—Estimated Fission-Product Source Requirements (*10*)

	Assumed dosage (rep)	*Treatment rate (lb/hr)*	*Utilization of gamma radiation (%)**	*Required size of source (curies)*	*(watts of gamma, assuming 0.75-Mev gamma)*	*(watts total fission-product power†)*
Liquid sterilization (e.g., milk)	2×10^6	9×10^2	12	3.4×10^6	1.5×10^4	3×10^4
Canned meat sterilization	2×10^6	6×10^3	32	8.3×10^6	3.6×10^4	7.2×10^4
Insect control in grain	2.5×10^4	8×10^4	15	3.5×10^6	1.6×10^4	3.2×10^4

* The utilization efficiencies are not necessarily for optimum designs so that the efficiencies should not be taken as any indication of the relative ease of doing any of these treatments. These are merely examples of per cent utilizations for assumed geometries.

† Assumed that beta equals gamma. Power is used hereafter as the measure of fission products, because it is believed to be of more value to the engineer than curies, particularly when considering the relation of fission products to reactors and processing.

There would be a small delayed neutron flux, but preliminary studies indicate that this can be shielded with such materials as boron or cadmium without appreciably affecting the gamma intensity (*12*). A slightly longer residence time in this external circuit also would eliminate these delayed neutrons. In addition, the materials proposed for treatment are mostly organic, and the neutron absorption cross sections of the components, principally C, H, O, N, S, P, and Ca are fairly low, which is favorable for control of activation.

Radiation by this scheme lacks mobility just as the reactor core does. Presumably, one might consider making such an irradiation fluid reactor mobile, but offhand this appears unattractive, particularly because of the problem of reactor coolant at the irradiation site and the improbability of producing and disposing of power.

If one had a two-region reactor with the fertile material fluid, circulating the fertile material and utilizing its radiation is a possibility. This radiation would be from product intermediates, like Pa^{233} and Np^{239}, and the relatively small amount of fission products.

Use of the radiation from Pa^{233} and Np^{239} in discharged solid units or as separated materials is a possibility, but their short life might limit such use.

Enhancement and specific radiation by neutron activation. Certain isotopes absorb neutrons to yield products decaying with gamma or beta emission that might enhance the radiation. To result in any appreciable amounts, the irradiated material must have both a satisfactory neutron absorption cross section and sufficient isotopic abundance, and the product must have a short life and yield an intense radiation on decay. Such irradiations could be by incorporation of the additive in a fluid fuel system or as a separate fluid loop in any reactor.

This might be particularly desirable if there should prove to be certain uses, such as chemical reactions, which need a specific gamma or beta radiation available from such special irradiations but not from fission products. However, the author knows of no significant specific uses of this sort reported to date. Utilizing any beta radiation from a fluid reactor would be difficult, but use of very thin metallic walls or possibly plastics, might be a faint possibility, although hazardous.

This enhancement amounts to trading a neutron for a specific radiation. Energywise it is a poor trade (184 Mev and fission products potentially from fission (*11*) versus a few Mev from radiation), but a specific use might justify it.

Future Reactor Complexes

The future fission-product radiation potential varies with the type of reactor considered. Four possible types of future reactors are considered.

Solid-system power reactors. Consider the industrial power picture when reactors operate commercially to produce power. Assume a 500-Mw heavy water moderated, light water cooled, natural uranium, power reactor of the type studied by the Pacific Gas and Electric Co. and Bechtel Corp. industrial power team (*13*). (This reactor is chosen as an example instead of those reported on by the other industrial power teams, because the design data is presented in a more useful fashion for the purposes of this article.)

We will further take W. H. Zinn's assumption of a 1% consumption of the total uranium before processing for this general type of power-only reactor (*14*). Using the conversion of 1 gm of material fissioned being roughly equivalent to 1 Mw-day of energy (*15*), this 1% consumption or "burn-up" converts to about 10,000 Mw-days per ton of fuel. On the basis of the estimate of 60 tons of natural U for this 500-Mw reactor (*13*), the reactor irradiation time to discharge of fuel is 1,200 days, or say 4 years considering that the reactor will probably not operate 100% of the time (*16*). Let us make the further assumption that 6 months is the total time for shipment to a processing plant, cooling, processing, packaging fission products, and shipment to the user. With such an operation, each batch of discharged fuel could yield about 0.5 Mw ($10^{-3} \times 500$) of fission-product power. This would decay to roughly 0.05 Mw when another batch is available in 4 yr (*17*).

The total installed capacity of electric utility generation plants in the U. S. including steam, internal combustion, and hydro, was about 75,000 Mw in 1951, having a doubling time of something over 10 years for the last 30 years (*18*). Let us assume that by 1970, the installed U. S. electric power capacity is 200,000 Mw and that reactors are furnishing 5% of this. At a 25% efficiency of reactor power to electric power (*13*), roughly 80 of these 500-Mw reactors would be required. This means that somewhere between 40 and 4 Mw, or possibly 15 Mw average, of separated fission-product power, less processing and packaging losses, would be available. This is not

a very large amount compared to the requirements for the uses suggested.

Fluid reactors. Solution fluid reactors producing Pu, U^{233}, power, or combinations, offer the potentiality of immediate processing on a continuous basis to give the freshest of fission products. Since only one fluid reactor of any size, the Oak Ridge homogeneous reactor, is known to have operated, the technology is probably in an early stage, and the processes and processing schedules of future fluid reactors are, therefore, conjectural. It may be possible to remove the gases, Xe, Kr, and I quickly from solution fluid systems; using these and their decay chains as a source has been suggested (*19*). Subsequent elements in the fission-gas decay chains are probably solids at ordinary temperatures, so that there is not continued mobility of such a source by pumping as with the original gases. No doubt certain fission products would be allowed to build up in the reactor system before any processing, continuous or intermittent, is begun.

Obviously, if these fission products are not removed immediately from the system—and it is highly unlikely that all of them will—then an external fuel stream of fluid reactors would have large amounts of available fission-product power that might be utilized. For a 500-Mw reactor, there is about 30-Mw of fission-product power at equilibrium (6% of reactor power) (*11*). After cooling for 10 sec, which, from conventional pumping experience, seems to be a reasonable average age in an external circuit, the fission-product power would still be in excess of 20 Mw (*17*). (Although determining this 20-Mw value requires some extrapolation of the decay heat curves, it is believed to be a reasonable value for our purpose. There is also some additional gamma radiation from delayed neutron fission.) Assuming half of the fuel system outside the reactor, then somewhere between 10 and 15 Mw of fission-product power could be available in an external circuit for such a reactor at equilibrium.

Aqueous systems seem more interesting for such external fuel stream irradiation, since nonaqueous systems such as fused salts and liquid metals probably would provide more self shielding. Also, light or heavy water would tend to moderate the delayed neutrons, thus aiding neutron shielding. There would be a problem in insulating the hot fuel stream to prevent heat damage or "cooking" of the treated material. (The heat from the radiation absorbed by the treated material is probably not sufficient to do damage, e.g., 2×10^6 rep is 4 cal/gm or about a 4° C temperature rise.) Practicability must be resolved by economic studies, and will change with developments in technology. Engineering might be very difficult. Also, such use of fluid reactors might not be very compatible with the production of power, Pu, or U^{233}.

Therefore, there is a choice of using the fission-product power of a fluid reactor in either an external circuit or as separated fission products, the total availability approaching 6% of the reactor's power. Because of the predominance of very-short-lived fission products and the time inherent in separations and packaging, it seems

The Fission-Product Future: Some Conclusions

1. Proposed large-scale uses of fission products would require megawatt quantities of fission-product radiation.
2. The principal reactor sources of such radiation are (a) separated fission products from used fuel, (b) external fuel streams from fluid reactors, (c) discharged solid fuel, (d) the reactor core itself.
3. A relatively large number of commercial solid-fuel reactors of certain present conceptions would be required to furnish megawatt quantities of fission products, if the fuel irradiation time is as long as has been indicated.
4. The fission-product radiation potential of fluid reactors is much greater than that of solid-system reactors for the same amount of reactor power, because: (a) fluid systems offer the possibility of essentially continuous removal of fission products integral with operation of the reactor, with the short decay time leading to very high activity; (b) external fluid fuel streams offer fission-product radiation in large quantity (practically free from neutrons).
5. As an example of the potentialities of fluid reactors, assume that problems in milk sterilization, such as taste changes, are solved. Then the whole milk consumed as fluids by Chicago, 1.4 billion lbs. in 1948 (*20*), could be sterilized with about 0.5 megawatt of absorbed gamma radiation (assuming sterilization processing two-thirds of each day or 67% capacity factor on the source). Further assume that the sterilization equipment is only 10% efficient in the absorption of gamma and that only half of the fission-product radiation is gamma, then roughly one-third (or 10 Mw) of the radiation potentially available from 500 Mw of fluid-reactor power would satisfy this Chicago requirement.
6. The trend to commercial reactors, either solid or fluid systems, probably will entail a trend to more compact processes on freshly discharged materials. This should yield relatively more separated fission-product radiation in a form more convenient for packaging than do present reactors and processes.
7. Present and near future costs of fission products are not necessarily any indication of the costs to be expected when reactors and processes of a new type operate on a commercial basis rather then for military purposes.

likely that the available separated amounts would be much less than for the external circuit idea. Shipment from the reactor site would be limited to longer-life materials.

Slurry fluid reactors are a somewhat different problem. Here the amount of radiation in the external fuel stream could be about the same as for a solution system. However, the processing characteristics, and, therefore, the separated-fission-product picture, are much the same as with solid-system reactors, since it is improbable that the fission products will get into the fluidizing media and offer the same possibility of continuous removal. The irradiation time for a power-type slurry reactor may approach the long periods mentioned for solid-system power reactors, so that comparably smaller amounts of separated fission products will be available.

It is seen that fluid reactors offer interesting possibilities for large quantities of fission-product radiation. A few large fluid reactors might have the potential capacity to furnish the megawatt scale sources needed for large applications.

Pu production reactors (solid systems). Limiting our consideration to economic reactors precludes a discussion of reactors producing only Pu.

However, some of the more obvious features of such a fission-product supply may be interesting. Most important is the fact that any such supply is subject to the uncertainties of the weapon demand. In addition, because of the desire to have Pu in weapons rather than as inventory in a reactor and to avoid loss of Pu by fission during long irradiations, it is obvious that irradiation and processing must be more frequent than with the power-only reactors discussed. This shorter irradiation time leads to somewhat more available fission-product power for a given amount of reactor power.

Assume that the needs for weapon Pu results in a 1.3-year irradiation time instead of the 4 years, based on the PGE-Bechtel assumption that fuel fabrication and processing costs would be reduced one-third by operating the reactor for power only (*16*). Considering a 500-Mw reactor and a 6-month time to source preparation, one would have about 0.25 Mw ($5 \times 10^{-4} \times 500$) every 1.3 years (*17*); this would have decayed to about 0.08 Mw ($1.5 \times 10^{-4} \times 500$) by the time another batch of fuel is available.

Another View . . .

. . . of the availability and use of radiation from reactors and fission products is presented by Bernard Manowitz in an article starting on p. 18 of this issue that considers possible applications of radiation in the chemical industry.

Mobile power reactors. With mobile power reactors, either fluid or solid systems, the fission-product radiation supply situation is probably similar to that of the solid-system power reactors. This is because it is likely that space limitations and practicability would preclude either use of radiation with the external fuel stream or integral continuous separation of fission products for mobile fluid reactors. Also, the tendency would be toward long irradiations to limit refueling.

The mobile use with the least limitations on size would be naval vessels; these might be natural uranium fueled, with a supply of fission products as discussed previously. Reactors using enriched material, e.g., airplanes, would have a shorter "burn-up" time because of the lack of generation of fissionable material. Further, weight limitations would tend toward high reactor power densities, i.e., power per weight of fissionable material, so that "burn-up" might be more rapid. Thus such enriched reactors would tend to have relatively higher amounts of fission products available than natural U, or even slightly enriched, reactors.

In any case, shipment of fuel to a central processing plant, entailing loss of fission-product radiation by decay, is probable. The supply of fission products might be subject to uncertainties from intermittent operation, particularly if military mobile reactors are involved.

Trends in Processes

Although processing has been discussed already in relation to specific reactors, a few general comments on trends as related to fission-product supply may be pertinent. Past processes have been primarily aqueous separations designed to recover fissionable material only, leaving the fission products as worthless wastes that are not particularly amenable to fission-product packaging. Any processing for commercial reactors must be justified by the value of the contained materials, U^{235}, U^{238}, U^{233}, Pu, Th and fission products, in relation to the cost of recovery and the cost of storage or disposal. The competitive producers of these materials are: mining and refining U and Th ores, isotopic separation of U^{235} to compete with Pu and U^{233} for enriched power reactors, and other sources of radiation, e.g., radium, accelerators, and Co^{60}, to compete with fission products. Therefore, the supply of fission products would be intimately related to the economics of many other activities.

Studies indicate that the cost of a commercial reactor power complex must be considerably less than present reactor complexes. With respect to processing, the cost of processing can be reduced by less frequent processing (perhaps none at all) or cheaper processes. In general, cheaper processes would be expected to involve fewer steps and increased concentration of materials, meaning less equipment of a smaller size. It would appear that the normal trend to commercial reactors is also toward processes giving cheap fission products in a useful form.

* * *

Any conclusions stated or implied in this article are those of the author and are not necessarily the official views of the Atomic Energy Commission.

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Van de Graaff Accelerators for Sterilization Use

Distinguishing between presently available electron accelerators of this type and possible machines of the future, the authors discuss costs and characteristics in the light of sterilization requirements

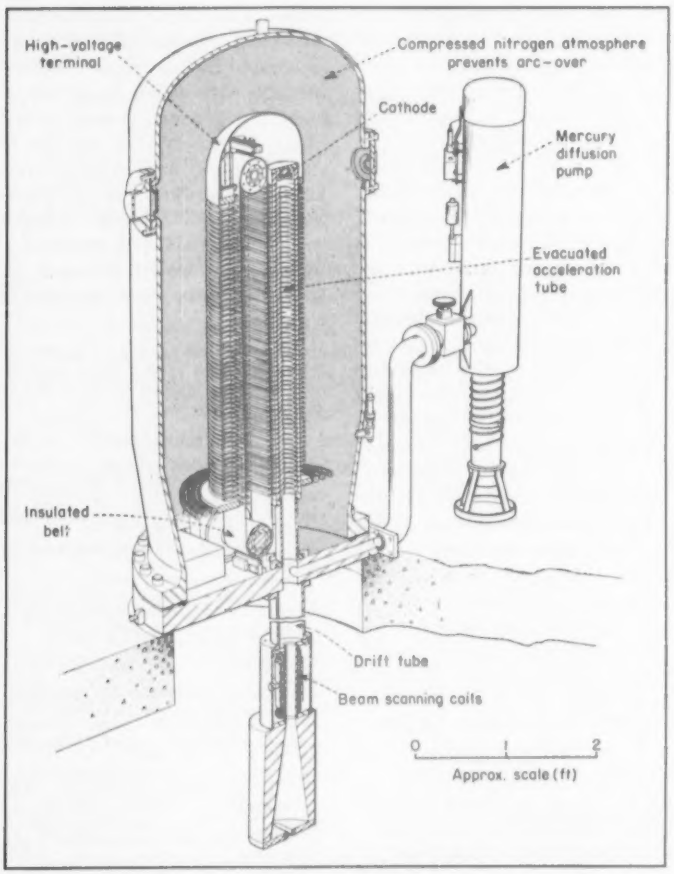

FIG. 1. Type AD 2-Mev Van de Graaff electron accelerator. Insulated belt moving rapidly carries charge to high-voltage terminal, creating electric field down length of evacuated acceleration tube. Both are surrounded by voltage generating column formed of series of equipotential planes separated by glass spacers. Electrons emitted from cathode are accelerated down tube, are diverted by beam-scanning coils that sweep beam from side to side to improve utilization efficiency, and pass through 6-in-wide aluminum window to strike material being irradiated

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AT LEAST FOR SOME TIME to come, machines will be the most readily available sources of radiation energy for industrial processing. This article is intended to summarize factually the present-day status of the Van de Graaff particle accelerator. Its state of development today will be described, and its capabilities and limitations discussed in the light of those possible industrial requirements that appear to challenge its capacity.

The basic operating principles of a Van de Graaff accelerator are illustrated in Fig. 1, a cutaway view of a 2-Mev unit (type AD). Over 40 units of this general type have been built commercially. Its major components, which include the voltage generating column, pressure tank, vacuum system, tube extension system and controls, are typical of all Van de Graaffs.

Aside from a control console, no other accessory apparatus is required. In addition to electric power (6 kva), the facilities required for an installation are water for cooling (2 gal/min), dry ice to cool vacuum-system vapor traps (10 lb/day), and local radiation protection.

State of Development

Table 1 summarizes the capabilities of available Van de Graaffs in terms of some criteria important in industrial

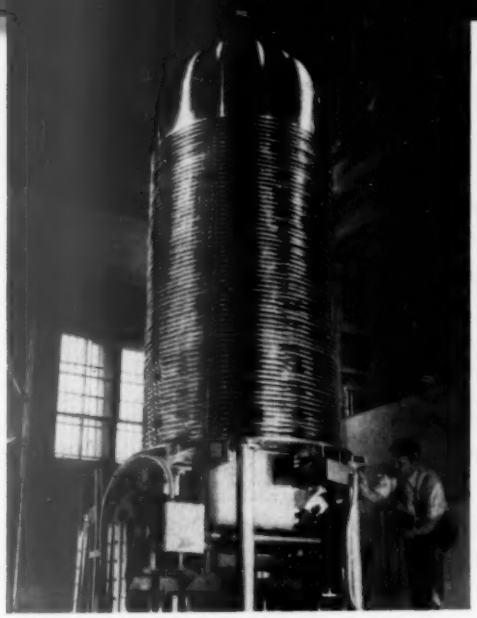

FIG. 2. Type FD 3-Mev Van de Graaff electron accelerator has total output of 12 kw

FIG. 3. Type JD 1-Mev Van de Graaff unit for research and pilot plant purposes has shielded irradiation chamber

processing. Existing 4- and 6-Mev accelerators have been omitted since they have, to date, operated as positive-ion machines only. Converted for electron operation, it is estimated that these higher energy machines would produce 3 to 4 kw of electron power without extensive modifications.

Figure 2 shows the 3-Mev type FD accelerator rated at 12 kw in continuous operation. Beam power output of 18 kw has been obtained from this equipment, which was designed as a prototype for high-volume sterilization applications. It is the most powerful source of electron radiation energy in existence.

The newest addition to the family of Van de Graaffs is a 1-Mev unit (type JD, Fig. 3) designed specifically as a research accelerator. Its useful output exceeds 1×10^6 gm-rep/sec, sufficient to process enough samples for most research purposes. To minimize installation requirements, this unit has its own radiation chamber that eliminates the need for other local shielding.

Production-Line Use

While the ultimate industrial requirements for ionizing energy are not fully known, probable requirements can be fairly well characterized:

1. It seems likely that many applications will involve the processing of large quantities of material. Therefore, considerable amounts of power will be required.

2. From a cost standpoint, this power should be utilized efficiently.

3. Most applications will require ionization in depth, and many will demand the penetration of comparatively thick products or containers.

It is of interest to compare the capabilities and limitations of the Van de Graaff accelerator in the light of these requirements.

Power output. The power output of any electron accelerator is simply the product of its average voltage times its average current. Since both average and peak voltage and current are equivalent in the case of the Van de Graaff, power output is simple to calculate, the power output of the Type AD generator (Table 1) being 500 watts.

Generator output in terms of the amount of material that can be processed in a given time can be calculated by relating power output to the electron dosage required: 1×10^6 rep = 3.8 kw-sec/lb. A 3.8-kw machine is capable, therefore, of delivering a dose of 1×10^6 rep to 1 lb/sec, or a sterilizing dose (2×10^6 rep) to 1 lb every 2 sec (assuming 100% efficiency). It should be noted that the treatment rate of a given accelerator varies in inverse proportion to the dosage required. Whereas an accelerator delivering 3.8 kw to the product can completely sterilize 1,800 lb/hr, it can deliver a deinfestation dose of 40,000 rep to some 90,000 lb/hr.

As is shown in Table 1, the power outputs of available Van de Graaffs range from 25 watts to 12 kw, the latter power capability representing a theoretical treatment rate (at 100% efficiency) for sterilization in excess of 5,000 lb/hr. Actually, of course, it is not possible to utilize the beam with 100% efficiency in practice. The efficiency with which the beam can be used is a major factor affecting both product treatment rate and cost.

Efficiency. Efficiency of beam utilization is limited by the intensity distribution of the beam in the product, both in depth and laterally. Figure 4 shows the distribution of ionizing intensity of a monoenergetic electron beam in depth, a pattern that holds true for all voltages above 1 Mev that might be utilized for sterilization pur-

TABLE 1—Capabilities of Available Van de Graaff Accelerators

Type	*Operating voltage (Mev)*	*Current output (ma)*	*Total power output (watts)*	*Beam utilization efficiency (%)*	*Penetration in water (cm.) (Double irradiation, 60% ionization level)*	*Approximate cost ($)*
JD	1.0	0.025	25	25–35	0.3	18,400
AD	2.0	0.25	500	25–60	1.6	56,200
FD	3.0	4.0	12,000	25–60	2.0	400,000+

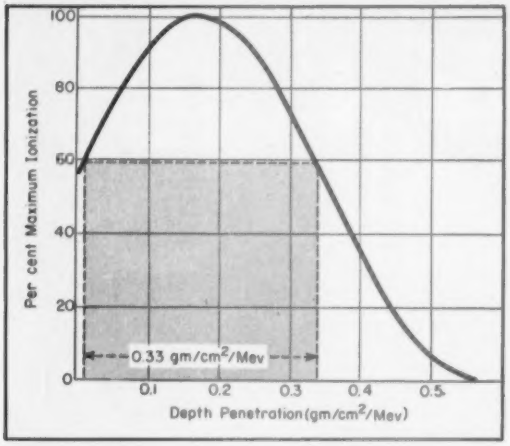

FIG. 4. Ionization intensity vs penetration for single irradiation from one side

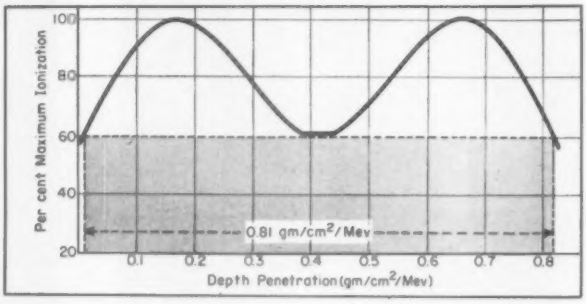

FIG. 5. Ionization intensity vs penetration for double irradiation

poses. The shaded area in the rectangle under the curve represents the maximum energy that can be efficiently used when equal dosage must be delivered to both top and bottom of the product. While the requirement for approximately equal entrance and exit doses may not be necessary for some industrial applications, it is almost certain to hold for most sterilization applications.

In the example shown, that of irradiation of the product from a single side, the maximum efficiency of utilization is 50%. Ionization intensity above the area of the rectangle is wasted in overdosing the center of the product. That to the right of the rectangle passes completely through the product and is not absorbed. This efficiency factor can be increased to about 70% and the overdose minimized, by irradiation of the product from two sides. The ionization curve for this double bombardment is shown in Fig. 5.

The lateral distribution of ionizing intensity across a stationary electron beam also limits utilization efficiency where it is necessary to deliver energy across the entire product width. The characteristic lateral distribution pattern of the electron beam is shown in Fig. 7, where the maximum possible efficiency is again approximately 50%.

This factor can be markedly improved by a technique applicable to monoenergetic electron beams. This is the function of the electron beam scanner (Fig. 1). Use of scanning permits the efficiency of lateral dose distribution to be increased to more than 90%, a typical pattern being that shown in Fig. 6. Scanning not only increases product treatment capacity significantly but also nearly eliminates the lateral overdose applied to the center of a product by a stationary beam.

Under the most adverse conditions—i.e., when a given minimum dosage must be maintained in all parts of the material being irradiated—the total efficiency of beam utilization can be summarized as follows: For a stationary beam, irradiation from one direction gives an over-all efficiency of 50% × 50% = 25%; from two sides, 70% × 50% = 35%. For a scanned beam, the respective efficiencies are 50% × 90% = 45% and 70% × 90% = 63%. These efficiency factors, when applied to the total power outputs of available equipment, give the treatment rates of Table 2.

Penetration. Limited penetrating power is the severest limitation of all present-day accelerators. In general, penetration of an electron in matter is directly proportional to its energy, or accelerating voltage, and inversely proportional to the density of the material being irradiated. The total range of electrons in matter of unit density is approximately 1 cm at 2 Mev, or, expressed differently, 1 gm/cm². For those applications requiring equal entrance and exit ionization levels (Fig. 5), useful penetration is 0.33 gm/cm²/Mev or about 0.66 cm at 2 Mev. Since the entrance dose (and by definition the exit dose) is 60% of the maximum ionization level, this distance is termed the penetration range at the 60% ionization level.

Where double bombardment is used (Fig. 9), this penetration range can be increased about 2½ times to 0.8 gm/cm²/Mev. This disproportionate increase occurs because the "tails" of the ionization curves are additive; it is a function of the slope of those curves.

The total and effective penetration capabilities of the three types of Van de Graaff electron accelerators are compared in Table 1 under conditions of double bombardment.

The fact that the penetration of the electron in matter is finite rather than exponential, has some definite advantages for surface treatment applications. Electrons can be readily used to irradiate a fraction of a centimeter of product surface to reduce or eliminate surface bacteria or to inhibit mold growth. Such applications appear promising for a number of reasons. A number of types of food spoilage are primarily surface phenomena. Low

TABLE 2—Effect of Efficiency on Output and Possible Treatment Rates

Accelerator type	*Effective output for equal entrance and exit dose (watts)*	*Treatment rate (lb/hr)*	
		Sterilizing dose (2×10^6 rep)	*Deinfesting dose (4×10^4 rep)*
JD (1 Mev)	8¾	4	200
AD (2 Mev)	300	142	7,100
FD (3 Mev)	2,400	1,130	56,500

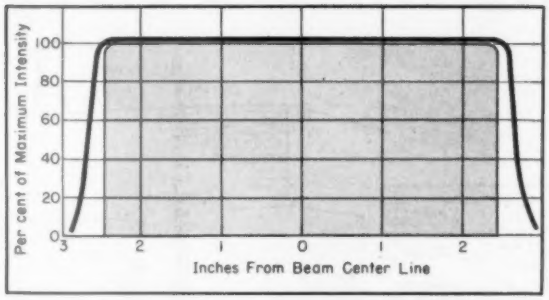

FIG. 6. Distribution of intensity across a 5-in. container with a scanned beam

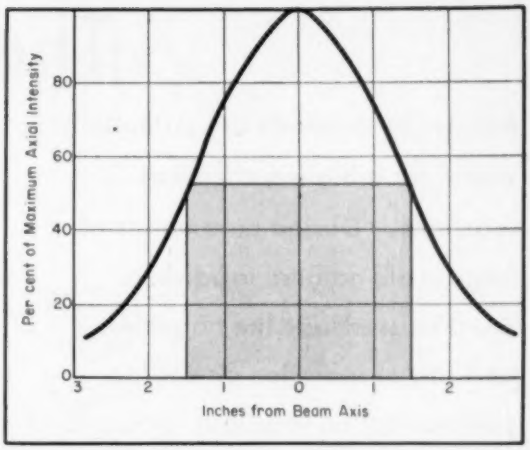

FIG. 7. Optimum distribution of intensity across a 3-in. container with an unscanned beam

voltages can be used to reach the necessary depths, and the production of such side effects as undesirable taste change, therefore, is limited to a small percentage of the total product volume.

Nevertheless, where thick products or containers are to be processed, limited penetration creates the need for high accelerating voltages. About 10 Mev would be necessary to treat a 3-in.-diameter can using double bombardment techniques. Where changes in packaging methods are considered impractical or uneconomical, higher energy accelerators will, therefore, be required. It is felt by the writers that design of such equipment is principally an economic problem.

Costs and the Future

A glance at Table 1 indicates that capital costs increase as voltage and output increase. Voltages substantially higher than 3 Mev are perfectly feasible today by means of simple conversion of the 4- and 6-Mev positive-ion accelerators to negative operation. The cost of this modified equipment should range between $250,000 and $350,000 as long as power output requirements are in the range from 3 to 4 kw. Still higher voltages or outputs would involve increased capital costs.

It appears possible, therefore, that interrelated cost and technical factors will limit the useful range of the Van de Graaff below the maximum voltages actually attainable with this type of accelerator. At the present time, this limit is not known, but it appears likely to be between 6 and 8 Mev.

Obviously, these capital costs, especially those associated with the larger machines, are much higher than would be the case if any important volume of production existed. There is no question whatsoever that significant improvements can and will be made.

For applications requiring considerably higher voltages, accelerators of the indirect type will probably be required. An example is the microwave linear accelerator from which several hundred watts have been attained, and which is today capable of developing voltages far higher than will be needed for most conceivable industrial needs. One unit, now being built by Stanford University and High Voltage Engineering Corp., for experimental electron cancer therapy at Argonne Hospital in Chicago, will be controllable in energy from 10 to 50 Mev. As more powerful microwave sources are developed, power outputs to satisfy the most difficult requirements may become available.

Within the voltage range of the Van de Graaff, the cost of obtaining adequate power for high volume industrial processing does not appear to be particularly formidable. The smaller Van de Graaffs are proving well adapted to low quantity production techniques and appear to be well suited for installation on conventional processing lines. Higher outputs can be expected from industrial units, and multiple units may prove to be more economical for many purposes than will large very high power single units. This seems especially true where processing is normally carried out on a number of filling or packaging lines.

A brief analysis of the costs of sterilization, using today's Van de Graaff, indicates that the cost per pound of product varies widely depending on the equipment required for the application and the dosages to be used. Such costs today, including a 5-year amortization schedule, operating, maintenance, and power, range from under $\frac{1}{10}$ cent per pound for high-volume, low-dosage irradiation to 7 cents per pound for low-volume, high-dosage irradiation. Similarly, costs per operating hour range from $8.00 to $36.00, and costs per kilowatt hour from $3.00 to $20.00.

The major element in these costs is, of course, the initial capital outlay that must be amortized over the production volume. The cost of power consumed is negligible. Lower capital costs realized as a result of increased production will, therefore, reduce these figures substantially. Electron power at less than $1.00 per kilowatt-hour appears to be a possibility within the next few years.

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The Industrial Future

Reactor by-products are potentially useful for initiating chemical reactions. Design parameters of large-scale gamma irradiators are discussed, and the potential and cost of radiation power for polymerization reactions is described

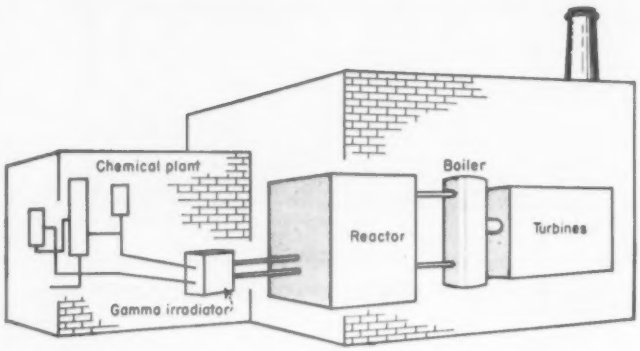

FIG. 1. Gamma irradiator uses short-lived fission gases at reactor

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THE FUTURE SUPPLY and cost of nuclear reactor by-product irradiators are such that they probably can be profitably used for only unique applications in industrial chemistry. This is indicated by an analysis of data on commercial atomic power and radiation-induced polymerization.

The present concept of nuclear reactors for atomic power is that heat energy supplied by the fission process would be used in a boiler supplying steam to a turbine-driven electric generator. At equilibrium the fission process releases about 200 Mev for each nucleus reacting. About 80% of this energy appears immediately as heat. The beta and gamma energies of the fission products amount to about 5% of the total energy of fission, and these are released gradually as the radioactive nuclides decay.

Fission-Gas Irradiators

While the fission-product energy is distributed among 20 different elements, about 20% of it is in the gaseous fission products xenon and krypton for about 1 min after fission. Thus, if the reactor fuel is a fluid from which these gases could be continuously stripped, transferred, and trapped in a gamma irradiator within1 min, about 1% of the total reactor power would be available for irradiations when the xenon and krypton are in equilibrium with their daughter nuclides. Initially, this equilibrium would be achieved in about 2 weeks, and the gamma irradiator would then maintain a constant power level.

Great penetrating power would be achieved because the short-lived fission gases and their immediate daughters have extremely energetic beta and gamma radiation. But the gamma rays are also energetic enough to interact with light nuclei and produce neutrons. While the neutron level will be low, neutron as well as gamma shielding would be needed.

The East River power plant in New York generates about 250 Mw of electric power; the power plant at Manchester, N. H., typical of many large industries, about 25 Mw. With a thermal efficiency factor of 4, nuclear reactors used to satisfy similar demands would require a heat supply of 100–1,000 Mw, providing a gamma radiation power supply of 1–10 Mw in associated gamma irradiators of the type described (Fig. 1).

One disadvantage of such gamma irradiators is that they would necessarily be located at the reactor site.

Mobile Gamma Irradiators

Periodic processing of spent reactor fuels to obtain the relatively long-lived fission products would make mobile gamma irradiators possible. The amount of fission-product power available as a function of time of burnup of fuel and time of cooling of spent fuel before fission-product extraction and use can be calculated from the Borst-Wheeler curves (*1*). Assume 180 days of burnup time, 100 days of cooling for short-lived mobile fission-product power, and 1,000 days of cooling for long-lived fission-product power. Then there would be 0.7 Mw of short-lived fission-product power and 0.03 Mw of long-lived fission-product power available from a 1,000-Mw nuclear reactor every six months.

When we consider that mixed fission products are available from all sorts of nuclear reactors, it seems reasonable that there some day will be a supply of 10-kw mobile gamma irradiators. Likewise, when we consider that the scheme described is just one way in which the gamma power of a nuclear reactor can be utilized, it seems conceivable that a 1-Mw gamma irradiator can be built in association with a nuclear reactor.

Roughly one-half of the total fission-product energy is beta, the other half gamma. The gammas will be used to create ionization in the material to be treated. The betas, with much shorter ranges, will be absorbed in the materials of construction of the irradiator, and will dissipate their energy in the form of heat. This heat must either be used in the process or removed from the system.

Another important characteristic of the fission products is their half-life as a function of time. For short decay times the half-life of the fission products is approximately equal to the decay time. After 6 years of decay, however, the fission-product power is relatively constant (30-year half-life) at a small fraction of the initial power. After 100 days of cooling the fission-product gammas are no longer energetic enough to undergo a (γ,n) reac-

# of Radiation Chemistry

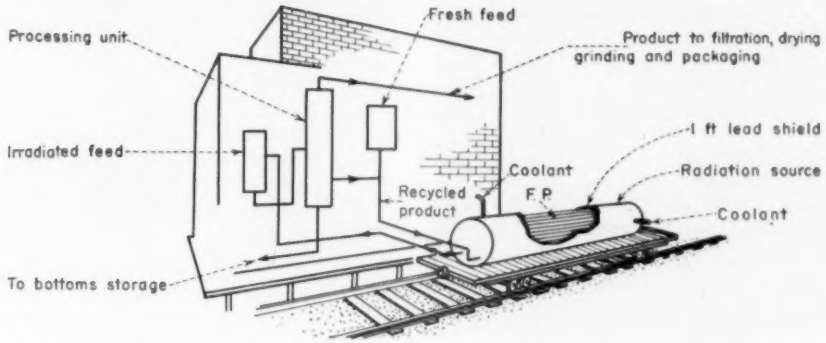

FIG. 2. Mobile gamma irradiator using processed fission products can be transported by rail

tion, so that no radioactivity would be induced and no neutron shielding would be required for mobile units.

Thus we can consider that there will be a mobile supply of short-half-lived materials that will require replacement or replenishment, say every 6 months, and there will be a smaller mobile supply of very long-lived materials that could be a relatively constant source of energy for periods up to 30 years.

One conceptual design of a gamma irradiator is that of a calandria with fission products on the shell side, materials to be treated on the tube side, shielded with 30–100 tons of high density material, all mounted on a railroad flat car (Fig. 2). The calandria and shield have a secondary cooling system for heat removal. Chemical reactions with high heats of reaction might require expanded cooling systems.

The unit is delivered to the processing plant, connected to the fluid lines of the material to be treated, used until the radiation power falls below desired levels, and is then returned.

Since the radiation from the fission products cannot be turned off, the fission-product irradiators pose a considerable personnel hazard problem. The irradiation devices must be so shielded that material to be treated can be easily exposed and removed from the radiation zone without any hazard to operating personnel.

Future Costs

To justify applied research and subsequent technological development in this field, it is relevant to inquire into the potential cost of fission-product power. One can have but tenuous hopes of approaching a realistic cost picture at so early a date, but, it is valuable to grope for cost limits.

In terms of design complexity, and, hence cost, the investment in an atomic power plant is likely to exceed that of an equivalent fission-product irradiator. Thus, atomic power costs might give an upper limit to probable cost.

Present investment costs for steam power are approximately $150/kw. Cisler has estimated (*2*) that future atomic power must not exceed $260/kw to compete eventually with steam power. Presumably this rate would apply to large power stations. Thus a 200-Mw power station (800 Mw of atomic heat power) would cost about $52,000,000.

We are, however, interested in much smaller scale plants. It seems reasonable to make use of the chemical engineering estimate that plant costs vary as the 0.6 power of the scale-up factor. On this basis a 1-Mw irradiator would cost $1,000,000, a 10-kw unit $60,000.

Polymerization Possibilities

With these ideas of what this device might look like and how much it might cost, let us look at what it can do in terms of pounds of product manufactured. Some experimental data on bulk polymerization reactions initiated by gamma radiation is assembled in the table on p. 20. These reactions are chemical chain reactions where the radiation must supply only the initiating energy. I have taken the yields at about 10–30% conversion and in further calculations have assumed that the conversion rate is linear with dose. This assumption is conservative in some cases, such as styrene and methyl methacrylate polymerizations, where the conversion actually increases with dose, and is optimistic in others such as solid-state polymerization of acrylamide, where there is a saturation effect.

For the purposes of this discussion, usage of a 1-Mw and a 10-kw gamma irradiator is considered. Assume that half of the total power is beta power, with all beta power wasted as heat in the core. Also assume that half of the gamma power goes into ionizing the materials being treated, and the other half into heat in the shield. To be consistent with the assumption that ¼ of the total power available is utilized, assume that the average intensity in both the 1-Mw and 10-kw irradiators is only 1 megaroentgen per hr.

To calculate the pounds per hour of product each of these irradiators can manufacture, it is next necessary to know the dependence of the over-all rate on the intensity of the radiation field. Three cases have been considered. One is based on the assumption that the over-all rate is independent of the field intensity, the second is based on the assumption that the over-all rate has a half-order dependence on the field intensity, and the third assumes that the over-all rate is linearly dependent on the field intensity. The only experimental evidence we have on the effect of field intensity is for the case of styrene polymerization, and here the rate seems to be linearly dependent on the field intensity. For the sake of conservatism, however, let

Experimental Data on Gamma-Initiated Polymerizations and Extrapolated Production Data

Polymerization reaction monomer	I *Gamma field intensity (r/hr)*	*Temp. (°C)*	*Pressure*	k, *yield (% converted/ megaroentgen)*	*Polymer molecular weight*	*Production capacity (lb/hr/Mw)* k = const.	k α $I^{1/2}$	k α I	*Ref.*
Styrene	250,000	−18	Atm	0.36	20,000	720	1,440	2,880	(5)
	190,000	30	Atm	2.2	70,000	4,400	10,000	24,000	
	250,000	72	Atm	10.4	300,000	20,000	40,000	80,000	
Methyl methacrylate	250,000	−18	Atm	10	~70,000	20×10^3	40×10^3	80×10^3	(5)
	250,000	30	Atm	32.4	~160,000	65×10^3	138×10^3	260×10^3	
	250,000	72	Atm	84	~250,000	170×10^3	340×10^3	680×10^3	
Perfluoropropylene	235,000	30	Atm	0.14	~600	280	573	1,190	(6)
Perfluorobutadiene	235,000	30	Atm	0.24	~325	480	985	1,970	(6)
Perfluoroacrylonitrile	250,000	30	Atm	0.10	—	200	400	800	(6)
Ethylene	120,000	30–80	Atm	0.1	—	200	574	1,670	(3)
	120,000	190	Atm	2.5	—	5,000	14,300	41,600	
Acetylene	120,000	30	Atm	0.13	—	260	565	2,170	(3)
Acrylamide	250,000	30	Atm	10–20	—	20–40,000	40–80,000	80–160,000	(6)

us use the half-order dependence as an assumption for intercomparison.

It should be pointed out that the experiments were performed as bulk polymerizations to obtain mechanism information, and that bulk polymerization, in most cases, probably would be impractical on a commercial scale. However, the rates of polymerization in suspension or emulsion polymerization would be faster, since the solvent would also contribute free radicals to initiate the polymerization. The faster rate would tend to balance the lower concentration of monomer within the reaction zone, so that extrapolations based on bulk polymerization should not be too unreasonable a guide.

The first two reactions are the polymerization of styrene and methyl methacrylate. Note that the molecular weight of the polymer varies directly with the ambient temperature rather than inversely as do polymerizations initiated by thermal or catalytic means. This phenomena will occur in all polymerizations initiated by radiation because the initiation rate will be temperature independent. The control of molecular weight over wide ranges is one unique way, then, in which radiation might be applied.

When we extrapolate the experimental rate information to production capacities in terms of pounds per hour per megawatt, we see that tremendous quantities of polystyrene and even more tremendous quantities of polymethylmethacrylate could be produced by a 1-Mw irradiator. However, if the 1-Mw irradiator is merely replacing a standard reaction vessel plus less than a ton of catalyst per day, the economic advantage of using an irradiator is not apparent. One interesting possibility is that of making an irradiator with field intensities high enough so that continuous polymerization could be accomplished rather than the batch operation used today.

The real driving force towards exploiting this technique, however, depends upon producing a material that has unusual and highly desirable properties. No physical tests have been made on the radiation polystyrene and polymethylmethacrylate, but these materials are not expected to differ much from the commercial variety.

The next three reactions indicate the possibilities of making entirely new polymers. Polyperfluoropropylene, polyperfluorobutadiene, and polyperfluoroacrylonitrile have never been made before by any other technique, to my knowledge. The yields were low and the polymers formed had low molecular weight, but these experiments do demonstrate that radiation is a powerful free-radical producer.

The polymerization of ethylene in a radiation field is indeed an interesting case, and might have a considerable commercial potential if our extrapolations are found to be valid, and if the polymer turns out to have reasonably good physical properties. Bretton and his coworkers (*3*) have demonstrated that polyethylene may be made at appreciable rates at temperatures in the range of 200° C and at atmospheric pressure. Since the present commercial process requires more highly elevated temperatures and extremely high pressures, this system certainly should receive further study.

Polyacetylene has also been made by the radiation technique. Should the polymer have interesting properties, it could be made at appreciable rates.

The solid-state polymerization of acrylamide is another example of a unique application of radiation. Acrylamide is a crystalline solid that melts at 84° C and can be polymerized by heating to 150° C. The table indicates that it will polymerize at an appreciable rate in the solid state in a radiation field. Solid-state polymerization could be a way of producing polymers with new properties.

A careful study of the mechanism of radiation-initiated polymerizations should point out other ways in which this tool may be used advantageously. For instance, using radiation, it should be possible to enhance branching during polymerization. As Charlesby's work with polyethylene has shown (*4*), it is possible to crosslink chains.

Other chemical reactions can also be induced by radiation.

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TABLE 1—Alpha and Li^7 Ranges in Air, Water, and Undeveloped NTA Emulsion

	α-particle			*Li^7*		
Energy (Mev)	*Air* (mm)*	*Water (μ)*	*NTA (μ)*	*Air (mm)*	*Water (μ)*	*NTA (μ)†*
0.5	3.4	4.4	2.2	2.9	3.8	1.8
1.0	5.7	7.4	3.6	4.1	5.3	2.6
1.5	7.9	10.2	4.9	5.1	6.6	3.2
2.0	10.5	13.6	6.6	6.1	7.9	3.8
2.5	13.5	17.5	8.4	7.1	9.2	4.4
3.0	17.0	22.0	10.6	8.2	10.6	5.1

* From Pollard and Davidson (*15*).

† Stopping power of NTA emulsion relative to air was taken as 1,600 from computations by Webb (*16*) and experimental observations by Lattes, *et al.* (*17*).

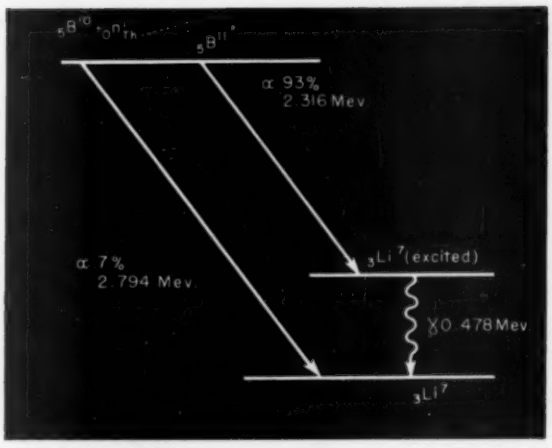

FIG. 1. Boron decay

Boron Detection in Tissues Using the (*n*,α) Reaction

Autoradiographic results obtained with the $B^{10}(n,\alpha)$ Li^7 reaction indicate that a method can be developed for determining boron distribution at low concentrations. Improved statistics should be attainable with more intense neutron fluxes

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IN RELATION to the larger problem of using neutrons for medical purposes, such as to irradiate neoplasms, experiments were made to determine whether boron could be detected in tissues at very low concentrations using the $B^{10}(n,\alpha)Li^7$ reaction.

Fast neutrons, while they can be directed at a local region of the body, have not been exploited for tissue irradiation because of technical difficulties in producing and controlling a pure beam and because an early study reported such discouraging results (*1*). Although thermal neutrons exist essentially as a neutron cloud (velocity of *circa* 2,220 m/sec at 0.025 ev), they can be pinpointed by creating in one area of the body a localized concentration of some element having a high cross section for thermal neutrons. The number of nuclear reactions occuring in that area would be proportionately increased over those taking place in the rest of the body. This differential dose would be most biologically effective if the nuclear reaction were to give rise to short-range, highly ionizing α particles.

Early biological experiments (*2–6*) confirmed the physical predictions. Most recently, while our experiments were in progress, Sweet and Javid (*7, 8*) described their initial studies on the use of this principle to treat brain tumors.

Isotopes having (n,α) or (n,f) reac-

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tions are Li^6, B^{10}, and U^{235}. Since only boron shows a tendency to concentrate in a body tissue, it has received most attention.

Natural boron is composed of 18.45% B^{10} and 81.55% B^{11}. The latter has a negligible capture cross section for thermal neutrons compared with the value of 3,900 barns for B^{10} (*9, 10*). Use of electromagnetically separated B^{10} in place of natural boron would increase the number of reactions and energy liberated nearly fivefold.

The B^{11} nucleus produced by capture of a neutron by B^{10} is unstable and disintegrates at once according to the schema shown in Fig. 1, with a Q value of 2.79 Mev (*11, 12, 13*).

Since momentum is conserved in this reaction, the available energy is distributed between the Li^7 nucleus and the α particles according to their relative masses (*14*). In 93% of disintegrations where an excited state of Li^7 is reached, the Li^7 nucleus takes $\frac{4}{7} \times (2.316)$, or 0.842, Mev, and the α particle, $\frac{3}{7} \times (2.316)$, or 1.47, Mev. Similarly in the 7% group, where the ground state is reached directly, the Li^7 nucleus takes 1.01 Mev, and the α particle, 1.78 Mev.

The ranges of these particles in air, water (tissue), and undeveloped NTA emulsion (Eastman) are shown in Table 1. These calculated ranges agree with the graphic data published by Herz (*18*) on unspecified nuclear track emulsions and on the experimentally observed range distribution of α particles produced by the $B^{10}(n, \alpha)Li^7$ reaction itself (*19*). The Li^7 ranges were computed by W. G. Pollard from $R_{Li}(E) = (\frac{7}{4})R\alpha(4E/7) + C$, where $R_{Li}(E)$ is the range of the Li nucleus in air at energy E; $R\alpha(4E/7)$ is the range of the α-particle in air at $(\frac{4}{7})E$; and C is an additive correction factor taken as 0.13.

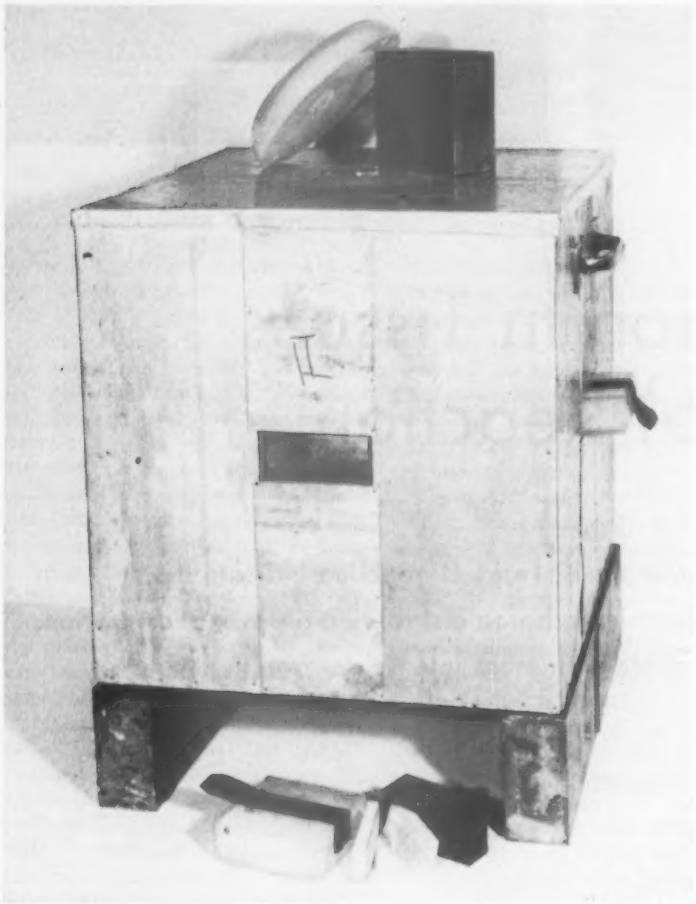

FIG. 2. Neutron-source box with plugs half out and slide box showing

Localization of boron. The most effective localization of boron must take into account both the differential distribution and the absolute numbers of B^{10} atoms present in the tumor.

Following large, toxicologic doses of the borate ion in both man and experimental animals, the borate and presumably all the boron showed a distinct tendency to concentrate in nervous tissue, liver, and skin, in that order; the nervous tissue contained 1.2 to 7 times as much borate as the liver (*20*). Smaller, more "physiologic" doses might result in quite different distributions. It has been shown in connection with studies on gallium that the size of the dose distinctly affects the differential distribution pattern of the metal (*21*).

In addition, some chemical form of boron other than the borate ion might give a different distribution. Another variable is that the distribution of a given ion in normal tissue may not hold for the neoplastic form of that tissue due to its peculiar metabolism or the changed "blood-brain circulatory barrier."

Procedure

The analytical method explored here was intended for studying concentrations below the limits of chemical methods. Also, by use of B^{10}, a reasonably good distinction can be made between the boron normally present in traces all over the body and the administered dose of boron.

Source arrangement. A Po-Be neutron source, 22 mm in diameter and 22 mm tall, was embedded in the exact center of a 38-cm cube of moderating paraffin sheathed with wood and cadmium. At the center of each of the 4 lateral faces of the cube, a rectangular box of Lucite, 3.5×8 cm (inside dimensions), extended into the paraffin so that the center of the inner face of each box was exactly 9.0 cm (radially) from the center of the source (Fig. 2).

Rectangular plugs of paraffin covered with Lucite were constructed to fit snugly into these caves. Each plug carried on its inner face a small hollow box of black Lucite into which were placed the 1×3 in. slides of tissue and the Eastman Kodak NTA plates (Fig. 2). When the loaded boxes were in place in the caves, the centers of the films were exactly 10 cm from the

center of the source. The slightly greater distance of the ends of the slide from the center of the source (maximum of 7.5 mm) appeared to be negligible.

Careful measurements using indium foil showed that all positions in each slide box were subject to the same neutron flux within the limits of accuracy of counting the activated foils (1.3%).* This is a result of the source not being a point and the preponderance of isotropic neutrons. The peak concentration of thermal neutrons from such a source in paraffin is approximately 10 cm radially (*22*).

A neutron flux of 5.76×10^3 neutrons/cm²/sec was obtained by an indium foil located at the positions occupied by the emulsion. Nearly 50% of these neutrons were in the epithermal region (3 ev or less) according to measurements with the indium sandwiched in cadmium. This value was corrected during the experiments in accordance with the 138-day half-life of Po^{210}. The exposure times during these studies were 8 to 14 days.

Except in the proximity of the caves on each of the four lateral faces of the cube, a $B^{10}F_3$ proportional counter detected negligible numbers of slow neutrons. Neutron-sensitive film badges and BF_3 pocket meters showed that this activity constituted no hazard.

The gamma activity at the surfaces of the cube measured 15 mr/hr using a Cutie-Pie calibrated against radium. The Po-Be mixture was the source of one 0.8-Mev photon per 2×10^5 α-particles, or 1 per 15 neutrons, and of one 4.5-Mev photon per two C^{12} nuclei formed, since one goes to the ground state via the first excited level. Other photons are emitted in the (n, γ) reactions with components of the moderator and shield. This level of γ-radiation, plus that from activation of nuclei in the glass slide and the emulsion, gave only a slight background fog. The use of plastic slides in place of glass definitely reduced fogging.

Sample preparation. Powdered B^{10} (98% pure) was converted to the borate by fuming HNO_3 and purified. This boric acid was dissolved in enough water to make a 5% (0.81*M*) solution after it had been adjusted to *p*H 7.4.

* The foil was calibrated by Dr. E. D. Klema, of the Oak Ridge National Laboratory, using an experimental reactor.

Each of four adult male mice received the equivalent of 1 mg of $H_3B^{10}O_3$/g body weight plus wash-in saline to make a total volume of injection of 0.025 ml/g body weight. Two mice were sacrificed 24 hr after injection and one each at 12 and 48 hr.

Ten tissues, including a blood clot, were removed from each animal and halved. One half was fixed in 4% formalin, washed, dehydrated, and embedded in paraffin using standard histological techniques. The other half was fixed and dried by the snap-freeze-vacuum technique, which, with the proper precautions, minimizes translocation and practically obviates leaching of the borate. Forty micron sections from the middle of each paraffin block were mounted on plastic microscope slides but not stained.

TABLE 2—Average Track Count per Field for Eight Preparations

$H_3B^{10}O_3$ concentration (%)	$\bar{x}$ tracks/field ± S.D.*
5	1.12 ± 0.17
5	1.16 ± 0.20
5	1.10 ± 0.16
5	1.05 ± 0.17
	$\bar{x}$ = 1.11 ± 0.18 (16%)
1	0.32 ± 0.10
1	0.28 ± 0.09
1	0.29 ± 0.09
1	0.30 ± 0.10
	$\bar{x}$ = 0.30 ± 0.10 (33%)

* Mean background α-track count was 0.07 ± S.D. 0.06 and should be subtracted to obtain net tracks per field.

By use of appropriate plastic and paper spacers, the tissue sections were mounted so that the section just cleared the surface of the emulsion. They were then packed in the light-tight boxes on the ends of the plugs and exposed to the neutron flux for 8 to 14 days.

The emulsion was developed in D-19 for 5 min and fixed in F-5 for 40 min. The tissue section was then realigned on its developed autoradiogram and the margins of the tissue outlined on the film using low-power magnification; artifacts, if present, were blocked out. These areas were then examined under oil immersion (970×) for α tracks.

Track counting. A reticle in the ocular was used to mark off 1,600μ^2, the unit area counted.

The α tracks counted were straight thick tracks between 2 and 5.59μ long. The minimum of 2μ was possible because the background fog was low enough to permit definition by use of three grains. Therefore, the great majority of α particles originating beyond the 6μ depth in the tissue would not have enough residual energy to register on the emulsion. Actually, however, the maximum "effective" depth, when worked out by reference to a series of geometrical approximations, proved to be 3μ. This results from the fact that the particle must enter the emulsion at an angle of between 0 and 60 deg to the horizontal; at greater angles, the grains are too superimposed to be distinguishable. This waste comprises nearly 20% of the hemispherical volume of emulsion theoretically available. Also, the deeper the point of origin of the particle, the more closely to this 60-deg maximum must it travel if it is to have enough remaining energy to activate three grains.

Tracks of protons and γ-rays were easily distinguished from the α tracks; Li^7 tracks were never clearly identified.

Blank preparations, consisting of drops of $H_3B^{10}O_3$ dried on the slides and of filter paper heavily impregnated with $H_3B^{10}O_3$, were used in preliminary studies of the tracks. Sections of gelatin containing the equivalent of 1% or 5% $H_3B^{10}O_3$ were used to simulate tissues having an even distribution of borate, a condition that probably does not exist *in vivo*.

In one large series of measurements, the background count of the emulsion was 0.07 ± S.D. 0.06 tracks per reticle field. Nevertheless, as a precaution against random contamination, background counts were made on each slide processed; approximately 20 fields located 5 mm beyond the margins of the tissue were examined.

The presence of paraffin used for embedding and sectioning appeared to make no difference in the background or track counts, so the sections were not cleared after being cemented to the slide.

All sections and slides of an experiment were handled and processed together to eliminate, as far as possible, variations caused by differences in temperature, timing, concentration of

TABLE 3—Tracks per Reticle Field of Tissues from Four Mice Sacrificed 12, 24, and 48 Hr after Injection*

	12 hr		24 hr		24 hr		48 hr	
Tissue	*form*	*f-d*	*form*	*f-d*	*form*	*f-d*	*form*	*f-d*
Blood clot	0.56	0.56	0.30	0.42	0.45	0.35	0.15	0.17
Brain		0.43		0.40	0.41	0.47	0.45	0.54
Heart	0.34	0.47	0.30	0.40	0.36	0.52	0.30	0.41
Kidney	0.48	0.41	0.30	0.55	0.47	0.60	0.25	0.42
Liver	0.61	0.57	0.40	0.43	0.35	0.51	0.33	0.43
Spleen	0.70	0.55	0.16	0.46	0.50	0.50	0.25	0.45
Pancreas	0.52	0.51	0.44	0.46	0.42	0.50	0.33	0.45
Lung	0.34	0.40	0.33	0.34	0.51	0.36	0.20	0.18
Muscle	0.30	0.45	0.30	0.43	0.33	0.42	0.36	0.50
Testes	0.47	0.51	0.20	0.46	0.38	0.44	0.36	0.29

* Tissues were prepared by either the formalin (form) or quick freeze-drying (f-d) fixation techniques.

solutions, etc. Similarly, all the NTA plates were stored together, and all of the exposed plates of an experiment developed and fixed at one time.

Results

The α-track counts on eight preparations of 1% and 5% $H_3B^{10}O_3$ in gelatin are presented in Table 2. Thirty reticle fields were counted for each preparation. At both concentrations, the maximum deviations from the mean were 5 to 7%, but the standard deviations of the means were high. This was largely an effect of the small numbers of tracks per field and the excessive skewness produced by zero counts per field. Consequently, the average number of tracks per field from the 1%-$H_3B^{10}O_3$ gelatin was not significantly different from the background, since the standard error of the difference between the two means (0.30 and 0.07) was ± 0.12.

The 5%-$H_3B^{10}O_3$-gelatin counts were significantly different; the standard error was ± 0.19. Where the tracks per field numbered 15 to 20, as with the B^{10}-impregnated filter paper, the standard deviations were usually 3 to 6% of their means despite the irregular surface of the paper. Even the highly irregular crystals of $H_3B^{10}O_3$ gave counts whose standard deviations were less than 15% of their means.

The average net number of tracks beneath the 5%-$H_3B^{10}O_3$ gelatin was only 4.5 times that of the 1% mixture. This was most likely due to the two series of slides having been put up, exposed, and developed separately. For quantitative work involving intercomparisons, a series of "standard tissues" having known concentrations of B^{10} would have to be exposed with each set of "unknown" tissues.

In Table 3 are the average numbers of α tracks per reticle field on the tissues of the 4 mice; the neutron-flux exposure was 10 days. Twenty reticle fields were averaged, except when the tissue was too small, when twelve were counted. Background counts average 0.07 ± 0.06 tracks per field.

In only seven of the thirty-seven instances did the track counts of the formalin-processed tissues agree with those of the frozen-dried. In the remaining, the frozen-dried tissues gave the higher count by a frequency of 5 to 1. This suggests that leaching took place during the wet-processing, even though the tissue section was taken from the center of the block and the counts were made on the central part of each section. However, as pointed out in connection with the gelatin sections, such low counting rates make it impossible to assign statistical significance to any of the differences in the table. Thus, the data of the table are intended to show merely that reasonable, consistent data can be obtained by the procedures described.

Discussion

The number of nuclear activations A, and therefore of α-particles, expected from a specimen presenting a surface of S cm² at a distance of r cm from the source is (*24*)

$$A = \overset{t}{F}\left(\frac{S}{4\pi r^2}\right)(t\sigma N)$$

where:

F = the thermal neutron flux of a point source. Here it is combined with $S/4\pi r^2$ in the indium foil calibration value for thermal neutrons 10 cm from the center of the source and is 5.7×10^3 n/cm²/sec. F is variable, although in this experiment it is constant, neglecting decay. F cannot be increased indefinitely because the background fog would also increase to the point of interfering with identification of the shortest tracks.

t = the duration of exposure in seconds. It is limited due to fading of the latent images and expediency: 8.64×10^5 sec = 10 days.

σ = the absorption cross section for the reaction with B^{10}. For the mixture of slow and thermal neutrons here, it is a constant of approximately 3.9×10^3 barns or 3.9×10^{-21} cm² (*9, 10*).

N = the number of target atoms of B^{10}/cm³ of the tissue. A dose bordering on the toxic was given, the equivalent of 1 mg of $H_3B^{10}O_3$ or 0.164 mg of B^{10} per gram of body weight, comprising 0.99×10^{19} atoms. Since the excretion is about 50% (*20*), there should be an average of 5×10^{18} atoms of B^{10} per gram or cubic centimeter of tissue.

Thus $A = 9.6 \times 10^7$ activations/cm³ of tissue in 10 days. But, of this number of α-particles, only a fraction were able to reach the film and produce an identifiable track. Accordingly, the following corrections must be applied:

1. Only those particles within 3μ (3×10^{-4} cm²) of the surface are able to reach the emulsion with sufficient residual energy to make a track.

2. Half of the α-particles formed move *away* from the film.

3. Only about 80% of the tracks are identifiable due to the vertical overlap of grains.

With these corrections, the film should have $(9.6 \times 10^7)(3 \times 10^{-4})(0.5)(0.8)$ or 1.15×10^4 tracks/cm^2. In terms of the reticle field, which is 1,600 μ^2, there should be 0.18 tracks per field.

This value is of the same order as those given in Table 3, although somewhat lower. The difference may be due to a higher neutron flux than that given, or to greater retention and segregation of B^{10} than was estimated. Also, it is possible that tracks of Li nuclei originating from some of the B^{10} atoms in the very superficial layer of tissues (cf. Table 1) may have been included, since even in the preliminary experiments Li^7 tracks were not distinguishable.

In any event, this correspondence between theory and experiment supports the belief that the method can be developed into a practical means of determining small amounts of boron. However, a more intense flux of neutrons obviously is needed. To be on a statistically comfortable basis, the neutron source should provide some 10^6 n/cm^2/sec without much further increase of photon flux. The photon emission of Ra-Be and Sb-Be sources make them completely unsuitable, and neither the carbon cave used by Conger and Giles (*25*) nor the water moderating column attached to the Oak Ridge reactor gave suitable neutron/photon ratios.*

The disadvantages of this technique are that it is time-consuming and track counting is tedious work, and it would be expensive in terms of B^{10} and equipment. However, if the experiments of Sweet *et al.* continue to be promising, the cost will be small in view of the need to know precisely what amount of what form of boron will give the highest tumor-to-other-tissue ratios at various times after administration.

* We wish to express our thanks to Mr. F. J. Muckenthaler of the Oak Ridge National Laboratory for his assistance in these preliminary experiments.

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Radioactive Tank Removed from NRX

PROBABLY THE LARGEST RADIOACTIVE OBJECT that has ever been handled, the aluminum tank known as the "calandria" has been taken out of the NRX reactor at Chalk River. Several tubes containing uranium rods burst within the 2½-ton tank Dec. 12 (NU, Jan. '53, p. 76), putting the reactor out of operation.

Under the direction of Gordon W. Hatfield, manager of industrial operations at Canada's atomic energy project, the extremely "hot" tank was lifted out of the 35-ft-high reactor structure during the evening when few employees were at the project. A U. S. group assisted in the removal.

Special equipment had to be designed and built at the project to examine remote parts of NRX and to remove the burst tubes. Viewing devices were built to determine which of the 176 uranium-containing tubes within the calandria had burst. Methods had to be devised to withdraw these before the calandria could be taken out.

Wearing protective clothing, including respirators, workers performed certain work within the few minutes allowed. Then other workers had to take their places.

Once the 176 fuel tubes had been removed, the radioactivity of the aluminum calandria in which they had been inserted was studied with probes. Then the carefully rehearsed removal operation was carried out.

The 10-ft-high, 8-ft-diameter tank was lifted out of the reactor structure with an overhead crane, swung over a canvas bag attached to a skid turned on its end against the side of the reactor, and lowered into the bag. The skid was then lowered into a horizontal position on the floor of the reactor building and pulled out of the building by a tractor. A grader was hooked to the skid to tow it through the project and to a burial ground outside the plant. The grader was the heaviest rubber-tired vehicle on the project.

The calandria was towed to a spot well away from the project but still within the 10,000 acres controlled by the project. There a bulldozer heaped sand over the tank to shield it. One end was left accessible for study.

Decontamination of the reactor structure is continuing.

Problems in Operating a Research Reactor

Operation of a research reactor presents three types of problems:

- **TECHNICAL.** Poison effects of fission products and in-pile experiments, induced radioactivity and heat production in experimental apparatus, and packaging of irradiated material must be considered
- **ROUTINE MAINTENANCE.** Differs from ordinary industrial maintenance only in the radiation hazard involved
- **PERSONNEL.** Most of the operating personnel would have an industrial rather than an academic background. Care must be exercised in selection and training of personnel

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OPERATION HISTORIES of large, well designed reactors seem to divide into at least two distinct phases: The first phase, a short, intensely vital period of the initial start-up and operation to verify the reactor's design characteristics. The second phase, the period of normal operation, in which the reactor fulfills its assignments.

The loading of the Brookhaven Reactor was started on August 13, 1950, and criticality was reached nine days later. By December 18, 1950, the reactor was available for routine operation for research purposes. During these sixteen weeks, the evaluation of the reactor was done in great detail on a 24-hr basis by a group of highly qualified scientists under the direction of L. B. Borst. This work consisted of performing experiments that were planned and documented well in advance of the completion of construction.

As soon as it became evident that all of the theoretical predictions had been verified, the operation of the reactor was tailored to meet other requirements. This phase change is probably more abrupt for the well understood, graphite moderated, natural uranium piles than for some of the more recent designs.

The problems of normal operation of reactors are obviously as varied as are the purposes for which reactors are built. Large research reactors have one important operational requirement, however, which is common to all of the large reactors operating today. This requirement is that the reactor operate seven days a week, 24 hours a day, with as few interruptions as possible; these interruptions being consistent with installing and removing experimental apparatus. This requirement characterizes the operation and maintenance problems as being similar to those of a continuous chemical process or of a steam plant operated for electrical power production. For this reason, most of the people actually engaged in the operation of the reactor have an industrial rather than an academic background.

At first sight it may appear difficult for the experimentalist and operator to meet on a ground common enough to expedite research problems. Unlike most other reactors, the Brookhaven reactor has no production commitments, and the operating staff is therefore dedicated entirely to the assistance of the research groups. The absence of production commitments is essential to such a cooperative atmosphere. The modus operandi, so far as possible, is established by the requirements of the research programs. Conflicts usually occur only with other research requirements, and whenever a priority must be established, the operators are involved only as necessary to make proper contacts.

The Brookhaven reactor operates about 90% of the time with the shut downs regularly scheduled on Tuesday

evenings for as long as necessary to install or remove research apparatus and to do maintenance. Every effort is made to maintain this schedule. Unscheduled shut downs interrupt experiments and may in some cases cause repetition of time-consuming measurements.

The problems of operation may be classified into three distinct categories: the immensely interesting technical problems, the highly routine maintenance problems, and the all-important personnel problems.

The achievement of proper balance of effort in these directions is in itself a problem. The tendency to expand efforts in the more interesting field at the expense of the more routine and unpleasant fields is understandable and establishes a requirement for specialists in each category.

TECHNICAL PROBLEMS

It is necessary to load the reactor with sufficient fuel to override all of the negative coefficients or poison effects that are brought about by the neutron flux present in the reactor at its full power level. In addition, a comfortable amount of reactivity to support in-pile experiments is desirable. Other problems associated with the experimental equipment are induced radioactivity, heat production through neutron and gamma reactions, and proper packaging of irradiated material.

Reactivity Control

Fission product Xe^{135} has an extremely high cross section for neutrons, and its formation is by far the most important of these negative coefficients.

From the following decay chain

$$Te^{135} \xrightarrow{2\ min} I^{135} \xrightarrow{6.7\ hr} Xe^{135} \xrightarrow{9.2\ hr} Cs^{135} \xrightarrow{2 \times 10^6\ yr} Ba^{135}\ (stable)$$

$$Xe^{135} + n \rightarrow Xe^{136}\ (stable)$$

it may be seen that when the reactor is operating, the equilibrium concentration depends upon the flux density (formation rate of Te^{135}), decay of Xe^{135}, and burnup (formation of Xe^{136} by capture), also a function of flux density.

When the reactor is shut down, the concentration of Xe depends upon the decay of I^{135} and Xe^{135}, the latter being the slower in decaying; therefore, depending upon the operating flux density, the concentration of Xe^{135} may be considerably greater just after a shut down. In a very high flux reactor, this concentration may conceivably be great enough to prevent restarting for a number of hours. A reactor operating at 6×10^{12} n/cm²/sec at a fairly high temperature does not have to maintain excess reactivity to override xenon growth after shut down for two reasons: first, this flux produces only a small amount of buildup after shut down, and, secondly, the loss of heat during shut down provides an additional amount of reactivity that is immediately available.

It is interesting to note that, after startup following shut downs of certain periods of time, the rods must be inserted to hold constant power because Xe^{135} burnup rate is greater than I^{135} decay rate when I^{135} is below its saturation value.

Temperature coefficient. The magnitude of the temperature coefficient of a thermal reactor depends primarily upon the temperatures of the uranium and moderator during operation. The negative coefficient is a result, among other things, of the diminishing value of the cross section of U^{235} at higher neutron temperatures.

The coolant of a reactor usually produces some effect on reactivity between low-power critical conditions and full-power condition. At Brookhaven, the coolant is air and the sign

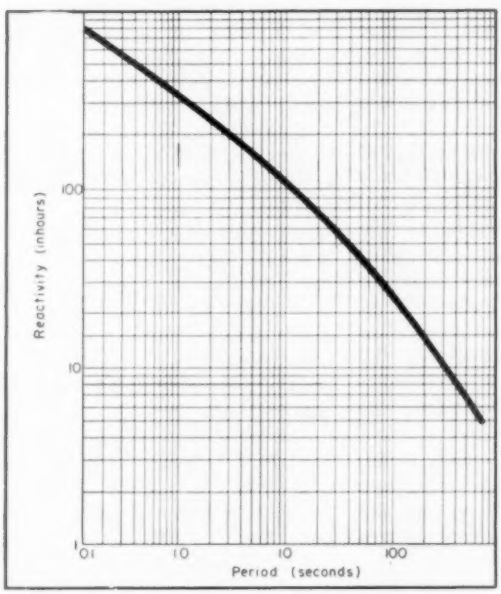

FIG. 1. Reactivity vs period found by withdrawing control rod a known distance

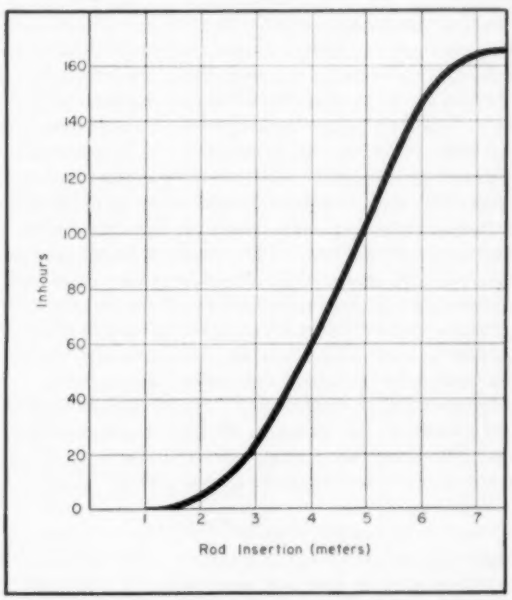

FIG. 2. Control-rod calibration curve (rod diagonal to horizontal plane through reactor)

INSTALLING in-pile equipment in Brookhaven reactor

EXPERIMENTAL apparatus at reactor face

of its coefficient is positive, since the coolant becomes less dense at the high-flow and temperature conditions. (Air is "sucked" through the reactor.) The change in density of N_2, with a 1.7-barn cross section, is primarily responsible for this effect.

Fortunately the magnitude of this positive coefficient is not great enough to constitute a hazard. An interesting fact is that, under certain conditions, power level may be changed from 20 to 24 Mw by increasing coolant flow without repositioning control rods.

The effect of weather changes on excess reactivity may vary with the particular pile. In the Brookhaven reactor, cooler air temperatures result in a cooler moderator in some areas of the pile, which might lead one to believe that cold weather causes a gain in reactivity due to the negative temperature coefficients. The converse, however, is the case, because the higher power levels afforded by the colder weather allow the growth of enough xenon to override the temperature effects and the net result is to decrease excess reactivity.

Fuel burnup. At neutron fluxes around 10^{12} n/cm²/sec, burnup of fuel occurs over a matter of years. Reactors operating in the 10^{14} n/cm²/sec flux range may require frequent fuel changes. Burnup problems are therefore proportional to flux and achieve great importance at higher fluxes.

Reactivity measurement. During the startup of a reactor, more than at any other time, the potential k, or total reactivity, must be known with fair accuracy. A reactor loaded for high-flux, high-temperature operation, after a long shut down, gains back all of the reactivity normally held by its negative coefficients. This gain, in some cases, may be sufficient to produce prompt critical conditions if control rods are removed too quickly. Even though in design the safety circuits are made adequate to prevent such conditions, it is well to predict the "critical position" of the rods so that control may always be maintained within the reaction time of the console operator.

It is desirable to hold the amount of excess reactivity the reactor has when it is operating at equilibrium to an absolute minimum. This is to say that at full-power equilibrium conditions, the control rods should be almost completely out of the pile. For a research reactor, this type of operation removes any perturbations to the natural flux distributions, and makes for more reproducible conditions. To accomplish this, all factors that affect reactivity must be continually evaluated. Among these, the ones more likely to cause frequent changes are due to experimental equipment changes and varying weather conditions.

A yardstick is necessary to conveniently make such measurements of reactivity. For most reactors this is done by measuring the amount of control rod inserted during operation at standard power level, with the temperatures of the structure and xenon formation in equilibrium. The effect of control-rod insertion on the reactor is usually not linear, since the rod enters from the edge of the structure where the flux is lowest and passes into the inner portions where the neutron flux is high. The yardstick is further complicated by the fact that its value or calibration changes as the fuel loading or poison loading is changed. The control rod must, therefore, be calibrated at each significant change in reactor loading.

There are commonly two units used to express reactivity of piles, and the calibration of control-rod length is expressed in these units. In the smaller size reactors, the term "Dollars and Cents" is used, a dollar being the difference in k between critical ($k = 1.000$) and prompt critical. (A pile is prompt critical when the reaction proceeds on prompt neutrons without depending on delayed-neutron contribution.)

The term used for the larger size reactors is "inhour," which is roughly defined as the amount of reactivity required to give the pile a period of one hour. To give some idea of the size of the units for a graphite pile

$$\$1.00 = 0.75\%\ k$$
$$289 \text{ inhours} = 0.75\%\ k$$

The use of two sets of units is often confusing. "Inhour" has been generally used for graphite piles. However, the tendency seems to be to use "Dollars and Cents" on smaller piles and critical assemblies.

A typical control-rod calibration procedure consists of bringing the reactor to critical at a very low power with the rod to be calibrated inserted far into the loading. By withdrawing the rod a known distance and observing the period with which the reactor power rises (time required for the power level to increase by e) the value of a known length of the rod is found by relating period to reactivity (see Fig. 1) from the equation*

$$\text{Inhours} = \frac{54}{T} + \frac{20.3}{T+0.62} + \frac{204}{T+2.19} + \frac{535}{T+6.5} + \frac{2036}{T+31.7} + \frac{787}{T+80.3}$$

If this procedure is repeated for various lengths of rod, a calibration curve similar to Fig. 2 may be obtained.

During operation, the position of control rods is continuously recorded, and the console operator plots inhours (excess reactivity of the pile) by totaling the value of all control rods inserted as indicated on the inhours versus rod-position calibration curves.

Determination of poison value. To determine the exact value of any poison inserted into the reactor, a typical procedure is to establish a critical position at low power, i.e., operate at constant power of 1 kw or less for a while and record rod positions. Next, shut down the reactor by inserting rods and insert the object in question into the reactor. Critical position is reestablished as previously described, and the difference in rod positions for the two conditions is equal to the poison value of the object, providing no other changes took place during the time consumed. The data may be badly "fogged" by changes in temperature and decay of xenon. Speed and isothermal conditions are therefore a requirement in these determinations.

In practice, these determinations of poison values of apparatus and accurate calibration of control rods are too time consuming and interfere with demands for continuous high-level operation to the extent that they cannot be done frequently. To follow the reactivity of a research reactor, therefore, one must be able to make educated estimates based on experience with the self-shadowing effect of high-cross-section materials and calculated values of poison.

Poison Prediction

The prediction of poison effects of research apparatus is probably 25% art and 75% science. The knowledge of chemical analyses, cross sections, weights, etc., allows one to make calculations that are helpful in that they usually are accurate to better than an order of magnitude. The discrepancies usually arise from the geometrical shapes of the higher-cross-section materials. Here, educated guesses often replace time-consuming experimental determinations.

Effect of Co^{60} shape. An example of the effect of geometrical shape of the in-pile apparatus on pile reactivity is found in Co^{60} kilocurie source production for Mr. B. Manowitz's program of obtaining high-intensity gamma sources. The cross section of Co^{59} for neutrons is 36 barns, which is large enough to make a small thickness of cobalt opaque or black to neutrons, depending upon the thickness of the specimen. If Co is to be irradiated in such a way as to produce the least effect on reactivity, its surface to volume ratio should be as low as possible, a sphere. Conversely, as is the usual case, if Co is to be irradiated for the highest possible Co^{60} formation, the surface to volume should be great. In practice, cylinders of Co with wall thickness of $\frac{1}{16}$ in. are irradiated. (The source may be further concentrated by selecting the diameters of the cylinders so as to allow them to be fitted into each other after irradiation.)

Neutron activation of such a 2-in. O.D., $\frac{1}{16}$-in. wall, 12-in.-long cylinder containing about 5 lb of Co was found by experimental determinations to be only 30% of the calculated amount, due to the local flux depression and self-shadowing.

The following gives some idea of the effect of 5 lb of Co on a pile:

Irradiated as solid rod: 6 inhours (value of a 16 in.^{3} completely black body)

Irradiated as a $2 \times 12 \times \frac{1}{16}$ in. cylinder: 25 inhours

At Brookhaven, the problem of control of excess reactivity is solved to a large degree by distributing cobalt through the reactor in such a way that a large number of small flux perturbations result. Such perturbations in a large-volume pile are difficult to measure. The demand for Co^{60} is sufficiently great to make this method of control much more economical than that of absorbing excess neutrons by boron in a control rod. Cobalt-60 production at Brookhaven is about 1,500 curies per month.

Induced Radioactivity

It is important that the induced radioactivity in research apparatus be predicted in advance, for the disposal of in-pile experiments may be the most difficult part of the experiment. A wide variety of shielded caves, reels, etc., are part of the equipment of a research reactor. However, these may not be effective if the materials of in-pile equipment are not chosen carefully.

The induced activity may be calculated subject to errors similar to those pointed out in discussing the Co^{60} problem and subject to additional errors due to lack of complete knowledge of the chemical constituents of the materials involved. Ordinary methods of chemical analysis do not point out the existence of minute materials that, depending on their cross section and half-life, may be more hazardous than the main constituent. Useful tables of isotopes, saturation values, decay schemes, etc., are available that are adequate, in most cases, for establishing the curie content of in-pile apparatus, providing its composition is really known.

The writer is not familiar with any method of calculating the amount of contamination that may be associated with removing in-pile experimental apparatus. High-level, air-borne contamination has been experienced when least expected. Elaborate precautions must be taken on all work of removing in-pile apparatus, and the assistance of an especially trained health physics group is usually required. A clear disposal plan for in-pile apparatus obviously should be made prior to installing the apparatus in the reactor.

Heat Production

Calculations must be made to back up estimates of heat production within the in-pile experimental material. This heat may be liberated from

* D. J. Hughes, et al., *Phys. Rev.* **73,** 111 (1948)

nuclear reactions involving absorption of neutrons, or from the absorption of gamma-ray energy.

Gamma heating of in-pile objects, especially well insulated heavy metals, is an appreciable effect which has trapped some of the best experimentalists. In-pile apparatus is also subject to damage from high temperature and radiation levels present in an operating pile.

The writer can recall one instance in which an elaborate in-pile furnace designed for controlling specimens at higher than ambient temperatures was redesigned to incorporate cooling after experience was gained with gamma heating.

Several values for gamma heating have been determined for some of the heavier metals. These, however, are not in close enough agreement to be relied upon. Whenever this phenomenon may be critical to an experiment, it is suggested that temperature determinations be made under the conditions of the experiment.

Packaging

Packaging of the materials to be irradiated in a high-flux reactor is a necessity established by the possibility of contamination of or by the material to be irradiated. The packaging is complicated by the heat production problems and the property of some materials to outgas during irradiation. Often the necessity of knowing the temperature in the vicinity of the object being irradiated further complicates the packaging problem by the presence of thermocouple leads, which must be passed out of the pile shield without producing radiation leaks.

Some specifications of packaging material are:

1. Container material should be such that specimen contamination is unlikely.

2. Container material should not "dust off" or be likely to contaminate handling tools and handling area.

3. Container material should have a short beta-gamma half-life, preferably beta with no alphas.

4. Container cross section for neutrons should be such that the container does not significantly shield the specimen from neutrons.

5. Container material must be available in shapes that can be mechanically worked and easily welded to provide "leak-tight" closures.

6. Container material must maintain its mechanical integrity under pile conditions.

Of the very few materials that meet these conditions, 2S aluminum for outer containers and quartz for inner containers are the most common.

The Canadians appear to be in a better position with respect to the procurement of very high purity Al. It has been reported that large amounts of 1S (very high purity) Al have been made easily available for use in the Canadian Atomic Energy Program. 2S Al is becoming increasingly difficult to locate, and delivery dates are unusually long for the amounts and shapes needed for research in the United States. The possibility of 3S or some other less pure grade of aluminum being substituted either in procurement or fabrication by someone who does not understand the problem completely is always a hazard. Aluminum shapes apparently lose their identity in storage if special precautions are not taken.

A comparison test is often made by irradiating a questionable piece of aluminum together with a piece of known 2S grade in the pneumatic tube facility for a few minutes. Half-gram samples are adequate, and the difference may be noted on ordinary survey instruments.

MAINTENANCE

The maintenance problem on a reactor is different from that of any ordinary industrial process only in that the additional hazard of radiation is present. The longer times required for surveying and decontamination increase the cost of such work. Aside from this, the procedures and methods are not uncommon in industry. Wherever possible, standard, time-proven products are used.

The relation of proper maintenance of a reactor's equipment to the safety of the machine and associated people is extremely important, and probably is much greater in proportion than the same relationship in industry. For this reason, preventive maintenance is practiced where feasible. About 70% of the instrument maintenance work is preventive in nature and includes mostly recalibration and testing components of the control system.

Heavy mechanical maintenance problems are about equal in magnitude and expense to the instrument portion of the work. At Brookhaven, the largest mechanical maintenance expenditure is on the cooling system and consists of the maintenance of ordinary swjtchgear, motors, fans, and pumps. This work is done mostly in the absence of radiation hazards. The cost of cooling the reactor, exclusive of maintenance, is by far the most expensive element in its operation. The cost of the 6,000-kw cooling load, on line 90% of the time, is about twice the cost of the salaries of the people actually involved in the entire operation.

ORGANIZATION AND TRAINING OF PERSONNEL

The operating staff must have the talents for making most of the calculations and estimates concerning items inserted into the reactor. In addition, theoretical physicists, metallurgists, chemists and other specialists must be readily available for consultation on nonroutine matters. As an operating staff gains experience, the actual time required of these specialists becomes so small that it is seldom necessary for them to actually be a part of the operating staff.

Since most scientists using a reactor for research purposes are specialists in their particular field of science, i.e., chemistry, biology, medicine, metallurgy, and physics, they are not usually experts on reactor operating problems. It therefore becomes necessary for the operating staff to carefully scrutinize all in-pile experiments to ascertain the following:

1. That the experiment must produce no hazard (radiation or otherwise) to the experimenter or other persons working in the vicinity.

2. That the experimental equipment must in no way endanger the usefulness of the reactor.

3. That there must not be any beams or scattered radiation that adversely affect other experiments in the vicinity.

This function is usually performed by the more experienced members of the staff, along with the Research Coordinator, a highly qualified man with research experience, who handles the scheduling and contacts with the research groups on these problems.

Probably the most important phase of operating a reactor is the selection and training of the staff that actually runs the pile. Chiefly among these are the engineers and operators on shifts, who operate process equipment.

Operating Staff Duties

At Brookhaven, the duties of the operators are probably more varied than at other larger sites. Fabrication of fuel, operation of pile equipment, general maintenance (including instrument maintenance), and handling of in-pile experiments are all duties of the operating staff. Since most of these associated operations never approach "production" levels, it is possible to conduct the entire operation with a total staff of less than 50.

A more complete description of the background, duties, and methods of training of these people is of considerable interest, since several similar organizations are rapidly coming into being.

To complete the picture of the job variety, an outline of the duties involved follows:

Operation of pile proper. This includes startup and control, data collection, calculation of heat balances, and charging and discharging fuel.

Cooling system. A 7,000-kw cooling system would require heat exchangers (air to water), cooling tower, and primary cooling fans (maximum size of rotating equipment: 1,500 hp, 3,850 rpm).

Water purification and storage. This involves an ion-exchange column and pumps associated with canal-water storage.

Fuel and sample fabrication. Operations involved are inert arc welding, hand torch brazing, induction brazing, and mass-spectrometer methods of leak detection and high-vacuum techniques.

Operation of 10-ton building crane and associated rigging.

Radiation survey work. This supplements regular health physics staff work.

In-pile equipment and service irradiations. This involves installation and removal of equipment.

Maintenance of heavy equipment, industrial-type instruments, and electronic instruments is done by specialists, who, for the most part, are members of the operating staff.

Training

About 75% of the Brookhaven National Laboratory operating organization do not possess college degrees. To assure the candidate's possession of adequate ability, a mechanical aptitude test is given. Experience has indicated that a cut-off point of 60% is advisable. This means that a score better than that of 60% of the college freshmen, who took the same test, is required.

For supervision of the operating staff, candidates with engineering degrees and at least five years of appropriate experience are desired. It is seldom that a supervisor can be trained adequately in less than one year.

A prerequisite to becoming an effective operating staff member is the gaining of self confidence in matters of radiation safety. This implies an ability to detect radiation hazards and to calculate safe working times as well as an idea of the possible effects of overexposure.

In most cases, approximately one month is spent with the health physics organization prior to reporting to the operating organization for pile training. During this period, all phases of personnel monitoring and radiation survey are covered, and the pupil must demonstrate his ability prior to moving on. This training is given to salaried engineers, as well as wage-roll operators.

Pile training is essentially "on the job type" training with manuals, models, and literature available. Progression is strictly on a merit basis, with demonstration of job knowledge and ability required for advancement.

Safety

In the operating of such a super critical machine, there is a potentially great hazard to equipment and, in some cases, to personnel when unsafe conditions exist. This hazard, however, is probably no greater than the average industrial chemical process.

Safety is stressed in many ways. The greatest effort in this direction is in the form of a manual of standard operating procedures, which covers all phases of the job.

If an operator consistently performs his work in accordance with these procedures, he may be considered a "safe" operator. If not, he is considered "unsafe," regardless of whether his infraction of the procedure causes an incident. Attempts are made to exceed the ultimate in the designing of safety into a reactor, but this has never, as yet, resulted in a completely safe machine.

As in all organizations involved in service work, the task of record keeping achieves importance. A careful study of record system must be made so that it operates as an asset to the program. Occasionally this work is pyramided to the point of becoming a great red tape burden to an entire operation. If it were not possible to cite examples, this point would not be made. For the Brookhaven National Laboratory reactor, no more than three people are occupied with these items, and this includes the work of billing all users of the pile's facilities, as well as computing Pu content of fuel and inventorying fuel storage.

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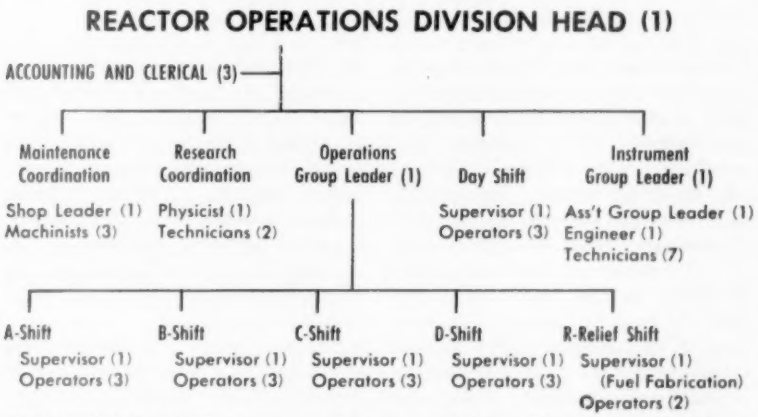
Research was carried out under the auspices of the U. S. Atomic Energy Commission.



ORGANIZATION CHART of Reactor Operations Divisions, BNL Reactor Department. During the regular work week, the day shift is considerably larger due to instrument maintenance and service irradiation requirements

Birmingham Nuclear Physics Conference

International progress in theoretical and experimental nuclear physics was reported in detail at a recent five-day conference. Summarized here are data on nuclear structure, reactions, and radii; high-energy scattering; heavy ions; light nuclei; theory of nuclear forces; and techniques

By JOHN K. BELING
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ONE OF THE LARGEST conferences on nuclear physics in recent years was held at Birmingham, England, July 13–18. The meeting was sponsored jointly by the University of Birmingham's departments of Physics, under P. B. Moon and W. E. Burcham, and Mathematical Physics, under R. E. Peierls. The attendance consisted of approximately 300 physicists from some 18 countries, and the discussions covered many phases of nuclear physics.

Program

The general pattern of the conference was to have the major topics developed in an introductory way by an authority in each field, followed by more detailed shorter contributions and discussion. *W. E. Burcham* (Birmingham) opened the meeting with a talk on energy levels of light nuclei. *B. H. Flowers* (Harwell) and *A. Bohr* (Copenhagen) spoke on nuclear models. A session on dense energy levels of highly excited nuclei was introduced by *K. J. Le Couteur* (Liverpool). *G. C. Wick* (Carnegie Tech) outlined the situation in the study of the nucleus by high-energy processes. The various mechanisms for the scattering of gamma rays were discussed by *P. B. Moon*, and *A. C. G. Mitchell* (Indiana) described techniques in nuclear spectroscopy. *S. Tomonaga* (Tokyo) surveyed the status of the meson theory of nuclear forces in a session held simultaneously with one on experimental methods. Other major topics included oriented nuclei and polarized beams of particles, introduced by *M. A. Grace* (Oxford), and a consideration of machine physics and the acceleration of heavy ions.

The wide range of topics discussed at Birmingham and the number of papers presented make it a particularly difficult conference to summarize, and it is hoped that the reader will make reasonable allowance for the subjective

nature of this report. It is understood that a fuller account consisting of notes taken by members of the Physics Departments at Birmingham will be available, at least to participants in the conference.

Birmingham Proton Synchrotron

A dramatic development in the progress of the Birmingham proton synchroton* occurred towards the end of the conference. This machine, designed for an eventual energy of 1 Bev, has been steadily achieving increasing proton energies as final adjustments and refinements have been made. A few weeks before the conference an energy of 250 Mev had been reached; during the conference this was raised suddenly to about 950 Mev.

The energy attained was inferred by noting how long the pulse induced by the proton beam on an electrode in the accelerating chamber persisted. The time at which the pulse disappears corresponds to a known magnetic field and energy. It should be remarked that Professors Moon and Burcham have been most restrained in their description of the synchrotron's progress, and have pointed out the possibility that the observed pulse corresponding to 950 Mev could be due to pickup from the r-f system. Nevertheless it seems probable that the 1-Bev energy region has now been reached in England.†

Nuclear Structure

The Birmingham conference gave evidence of further successes for the quasi-molecular model of nuclear structure developed by A. Bohr and B. R. Mottelson at the Institute for Theoretical Physics in Copenhagen. This treatment embodies features of both the liquid-drop and independent-particle models and describes the nucleus in terms of the coupling between collective surface oscillations and the motion of individual particles. It differs strongly from the independent-particle model in regions of the periodic table that are far from closed shells. There it predicts, for low-lying levels, a regular spectrum that arises from a type of rotational motion, whereas the independent-particle model cannot make such definite statements.

Bohr and Mottelson have received support for their model from the observed energy spacing of the levels of even-even nuclei in these parts of the table, and from the fact that electric quadrupole transitions between states of this type are found to be of the order of 100 times more probable than would be expected on a single-particle basis. Additional evidence for the collective model came from experimental work at the British Atomic Energy Establishment, Harwell, and the Institute for Theoretical Physics in Copenhagen.

Gamma rays from transuranic elements. *B. Rose* (Harwell) described measurements by himself and J. O. Newton of the energy of the gamma rays that follow the alpha decay of Cm^{242} and Pu^{238}. Here the nuclei formed by alpha emission are far from closed shells and would be expected on the Bohr-Mottelson picture to have levels of rotational character with even parity and successive spins $J = 0,\ 2,\ 4,\ 6\ldots$ Consequently, if the fourth level with spin $J = 6$ is excited by alpha decay it should return to the ground state, $J = 0$, by a cascade of three electric quadrupole transitions. According to the collective model, the energies of the successive radiations should be very nearly in the ratios 11:7:3.

Rose reported gamma rays from the decay of Cm^{242} of 160 ± 2, 102.8 ± 1.5 and 44.6 ± 0.5 kev, and three radiations of similar energies from a Pu^{238} source. All are in excellent agreement with the predictions of Bohr and Mottelson.

These accurate energy determinations were made by multichannel analysis of the size distributions of pulses produced by the gamma rays in Maze-type proportional counters filled with krypton or xenon. Rose and Newton were also able to obtain considerable evidence on the conversion coefficients, and in no case were these incompatible with electric quadrupole transitions.

Coulomb excitation of rotational levels. *T. Huus* (Copenhagen) described a clear experimental demonstration by himself and Č. Zupančič (Ljubljana) of a new technique in nuclear spectroscopy. A theoretical analysis by Mottelson indicated that excitation of nuclei by the Coulomb field of bombarding particles was possible and would provide results that could test the quasi-molecular model. This furnished the motive for the experiment, and Huus and Zupančič verified the occurrence of the effect by exciting levels in tantalum and tungsten with protons of about 2 Mev.

In this experiment the energy of the protons was so much below the Coulomb barrier that the cross section for the formation of a compound nucleus was only of the order of 10^{-38} cm^2. Since nuclear forces never come into play in this process, the cross section for excitation of a particular nuclear level by the Coulomb field of the incident particle can be expressed as the product of the nuclear matrix element for the decay of the level in question and a factor that can be calculated from the trajectories of the incident particles in the electric field of the nucleus. Since the collective model predicts that transitions between nuclear levels of rotational type should have large electric quadrupole matrix elements, these levels ought to be easily reached by Coulomb excitation.

One consideration in the choice of Ta^{181} for the experiment was that it is a nucleus where the Bohr-Mottelson model predicts a simple rotational spectrum, although one somewhat different from that of an even-even nucleus. Huus and Zupančič found gamma rays from the excitation of the 136-kev level in tantalum, and the cross section for emission of 136-kev gamma rays was about half a millibarn. They measured the energy variation of the yield and the angular distribution of the gamma rays and found good agreement with the theoretical predictions for Coulomb excitation. The collective nuclear model forecast the existence of a second level of rotational character at an energy 2⅔ that of the first, and, in fact, a search for radiation from the excitation of this state revealed a gamma ray at the expected energy. The cross section for the emission of the higher-energy gamma ray of measured energy 300 ± 10 kev was approximately 0.02 mb at a proton energy of 2 Mev.

Mechanism of Nuclear Reactions

Evidence concerning formation of a compound nucleus in nuclear reactions was presented by *H. H. Barschall* (Wisconsin), who described elastic scattering experiments that he and M.

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* L. U. Hibbard, NUCLEONICS **7**, No. 4, 30 (1950).

† The Birmingham team has since confirmed this by observations on protons emerging through the wall of the vacuum chamber.

Walt had done. Feshbach, Porter and Weisskopf had been able to explain the behavior of total neutron cross sections observed earlier by Barschall and associates, by treating the nucleus as a potential well. The well, however, had an imaginary term representing absorption to form a compound nucleus in some fraction of the cases.

Barschall's latest measurements were made to test the applicability of the new theory to angular distributions of neutrons scattered elastically by a large number of elements. The neutron energy chosen was 1 Mev, and a spread of about 100 kev was introduced to insure that many resonances of the compound system were excited.

The observed distributions were different from those predicted by the compound nucleus theory. They were, however, in reasonable accord with the new theory, although some ambiguity exists because the theory does not give the fraction of the compound nuclei formed that decays by elastic scattering. The present indication, therefore, is that the gross behavior of neutron reactions can be fitted with a potential of the form $V(r) = -V_0(1 + i\zeta)$ for $r < R$. For this fit a nuclear radius $R = 1.45A^{1/3} \times 10^{-13}$ cm is used. V_0 is taken as 19 Mev and the parameter ζ, which determines the absorption, is given the value 0.05, corresponding to formation of a compound nucleus in about 30% of the encounters.

Nuclear Radii

R. Hofstadter (Stanford) described instrumentation and preliminary results of high-energy electron scattering experiments done with the Stanford linear accelerator by H. R. Fechter, J. A. McIntyre, and himself. The magnetically analyzed electron beam of about 120 Mev was scattered from tantalum, lead and gold. Cerenkov counters used to detect the scattered electrons in a magnetic spectrometer were found to be particularly free from background.

The observed angular distributions were compared with those calculated assuming various nuclear charge distributions. Good agreement was obtained only by choosing a charge distribution that fell off very rapidly with radius.

This result provoked considerable discussion. Hofstadter emphasized its tentative nature and described limitations of the scattering calculations made so far that have involved use of the Born approximation. *R. E. Peierls* summarized the discussion by expressing willingness to accept from this experiment the qualitative fact that nuclei appear more concentrated centrally than previously expected. However, he pointed out the need for much more rigorous theoretical scattering calculations before the data could be interpreted definitely.

High-Energy Scattering Results

M. Snowden (Harwell) described measurements that he, Randle, Taylor, and Wood had made on the differential neutron-proton scattering cross section. They used a diffusion cloud chamber filled with hydrogen at a pressure of 20 atmospheres. Neutrons produced by bombarding a beryllium target with 170-Mev protons from the Harwell cyclotron were filtered through polythene to remove their low-energy component. Only proton recoils due to incident neutrons with energy greater than 100 Mev were used in the cross-section determination, and the effective neutron energy was 135 Mev. The cross section was measured between angles in the center of mass system of 22.5 and 160 degrees. A definite asymmetry with respect to 90 degrees was observed, the cross section tending to a value at 0 degrees of about half that at 180 degrees.

M. F. Kaplon (Rochester) gave an account of polarization experiments in proton scattering performed by W. F. Cartwright, C. L. Oxley and J. Rouvina. A double scattering arrangement was used, with the first scattering taking place from a target placed in the internal beam of the Rochester cyclotron. The polarization was detected by the azimuthal variation of scattering from a second, external, target.

Very large polarizations were reported for the scattering of the 240-Mev protons. The values were 49.8 ± 4% for a carbon scatterer and 20.6 ± 2.7% for scattering from hydrogen.

L. Wolfenstein (Carnegie Tech) pointed out that the polarization of proton-proton scattering reported from Rochester gives the first proof of the existence of tensor forces or spin-orbit coupling.

Heavy Ions

D. Walker and *J. H. Fremlin* (Birmingham) reported on the acceleration of various heavy ions, including C^{5+}, C^{6+}, N^{6+} and O^{6+}, to energies of the order of 10 Mev per nucleon in the 60-inch fixed-frequency cyclotron. High states of ionization were generally produced by the acceleration of singly- or doubly-charged ions to velocities at which they could be stripped to the final ionization by collision with gas molecules in the tank. Thus N^{6+} ions, for example, were obtained from initial N^{2+} ions that resonate at one-third the frequency of N^{6+}. The energy reached depends on the time spent by the particle in low states of ionization and the yield is relatively low; however, the method has the advantage of simplicity and flexibility.

Considerable work has been done at Birmingham on the bombardment of aluminum by N^{6+}. Eleven resulting activities have been identified. From a study of the yields of the possible reactions, a tentative hypothesis was advanced that the N^{14} nucleus is to be regarded as an almost unbound collection of nucleons in such processes. Thus it is considered to act like a sort of neutron-proton "buckshot." On this picture, true compound nucleus formation is improbable and the different reactions proceed by capture of a range of different groups of nucleons.

Light Nuclei

A considerable number of communications on the properties of light nuclei was presented at the conference. These included accounts of angular momentum and parity determinations and the effects of isotopic spin and charge parity selection rules on transition probabilities. The rapidity with which the latter type of data is accumulating was evident from a talk by *D. H. Wilkinson* (Cambridge), who discussed the experimental manifestation of these rules. He presented a table showing about 20 self-conjugate nuclei whose behavior could usefully be analyzed with the isotopic spin concept. The violations of selection rules of Wilkinson's listing, and other talks, could be due to Coulomb effects. Thus the Birmingham conference yielded no results demanding a charge dependence of specifically nuclear forces.

Theory of Nuclear Forces

S. Tomonaga (Tokyo) discussed the challenging situation in which the

meson theory of nuclear forces finds itself. He pointed out that close examination by several workers of the derivation of Lévy's promising new nucleon-nucleon interaction has removed much of its theoretical basis. However, it fits the low energy *n-p* and *p-p* data rather well.

E. M. Hafner (Cambridge and Rochester) discussed the experimental evidence below 5 Mev for the Lévy interaction. To preserve charge independence, i.e., equal singlet scattering lengths for the *n-p* and *p-p* systems, and the Yukawa well shape, a repulsion at short ranges of the Lévy type seems needed. Also, the Lévy potential does predict the magnitude of the scattering length quite accurately.

K. J. Le Couteur (Liverpool) presented a short semi-quantitative argument that illustrated the difficulties of the Lévy theory. He advanced reasons for believing that the meson theory of interactions between nucleons cannot be considered to involve only a single parameter in addition to the nature of the mesons and their coupling, but has a much more ad hoc character.

Techniques

A number of experiments were described that illustrated the refined nature of modern techniques in nuclear physics. These included an exposition by *M. A. Grace* of the subtle procedures used at Oxford to align and polarize nuclei at very low temperatures.

P. Huber (Basel) reported a measurement of the polarization of neutrons produced in the deuteron-deuteron reaction. The polarization was detected by the dependence on azimuth of the intensity of neutrons scattered from carbon. A polarization of 18 ± 7% was reported, and *R. Ricamo* (Zurich) described similar results obtained at the Swiss Federal Institute of Technology.

A. Berthelot (Saclay) described the successful use of a cloud chamber triggered from the pulse produced on an internal anode by electrons liberated by charged particles. The chamber was applied to studies of the frequency of ternary fission of uranium by slow neutrons. It was concluded that fission into three fragments all heavier than alpha particles occurred less often than once per thousand fissions.

D. E. Alburger (Brookhaven) gave a brief account of work he had carried out at the Nobel Institute on the transitions between excited states in Pb^{206} following electron capture by Bi^{206}. By use of several types of β-ray spectrometers to study conversion electron lines, and coincidence techniques, he was able to fix the location of 10 excited levels with a general accuracy of 5 parts in 10,000 and to establish a decay sequence.

Angular correlations. *S. Devons* (Imperial College of Science and Technology, London) discussed experiments that he and G. Goldring are performing on positron-electron angular correlations. Correlations were measured for the decay of the first excited state of O^{16} by pair emission and in other transitions where that process competes with electromagnetic radiation. He pointed out that an improvement that appears possible in the present accuracy, should make the experiment capable of distinguishing between the simple Dirac theory and the new radiation field interaction effects.

E. Heer and *T. B. Novey* (Argonne) discussed their experiments at Zurich on extra-nuclear effects that perturb the angular correlations of low-energy radiations. For the case of the γ-γ correlation in Cd^{111}, their group has established experimentally that the correlation may be influenced by anisotropic crystalline electric fields. Advantage has been taken of this effect to determine the electric quadrupole moment of the 247-kev first excited state of Cd^{111} which has a mean life of 10^{-7} seconds. The quadrupole moment has been determined to be 0.5×10^{-24} cm^2 with an accuracy dependent on that of a theoretical estimate of the electric field gradient at a lattice position in the indium crystal that contained the source.

Resonant scattering of gamma rays. *W. G. Davey* (Birmingham) described work that he and Professor Moon have done on the resonant scattering in mercury of the 411-kev gamma ray that follows the β^- decay of Au^{198} to Hg^{198}. The problem is to add to the gamma ray's energy the loss of energy in two nuclear recoils. This was done by mounting the Au^{198} source on the tip of a doubly tapered steel cone that was rotated at high speed to utilize the Doppler shift in the gamma-ray's frequency. Gamma rays resonantly scattered by mercury were detected by a scintillation counter operated with a narrow channel at high bias to discriminate against Compton scattering. Counting rate was measured as a function of rotor speed.

A resonance was observed at 1,700 revolutions per sec whose intensity corresponded to a half-life of $2.1 \pm 0.5 \times 10^{-11}$ sec for the 411-kev state in Hg^{198}.

Similar experiments where the recoil energy was provided by heating the source was described by *K. G. Malmfors* (Nobel Institute, Stockholm).

* * *

The opinions or assertions in this article are the private ones of the writer, and are not to be construed as official, or reflecting the views of the United States Navy.

New Units Speed Uranium Search

As part of its program to search out additional domestic sources of uranium, the Atomic Energy Commission has installed new radioassay units at 28 nonuranium mining facilities across the country. This sampling of base-metal ores at privately owned concerns is based on the frequent association of uranium with such ores.

Approximately 100 samples per day can be examined with the radioassay unit developed by the Instruments Branch of AEC's New York Operations Office. The 9 × 9 × 14 in. unit utilizes a "pill-box" halogen-filled Geiger tube that can detect uranium oxide at concentrations of 0.05%. A sample in a standard paper drinking cup is placed over the tube and counted for one minute.

Accumulated counts are displayed on a tricolored dial that indicates sample activity in terms of no radioactivity, little radioactivity, or activity at a level indicating uranium ore. Samples in the last category are sent to AEC for further analysis.

Monitoring of Water for Fission-Product Contamination

An annular liquid beta counter is described for measuring water contamination in the range 5×10^{-4}–0.5 μc/ml, with an obtainable accuracy of ±25%. A solid-source calibration jig obviates the need for liquid standards

By W. R. LOOSEMORE
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A LOW-VOLTAGE G-M COUNTER has been designed to monitor water contaminated with fission products in the range 5×10^{-4} to 0.5 microcuries/ml if the sample is diluted up to a maximum of ten times its original volume.

The counter is intended for use with a portable contamination meter (type 1092D) (*1*) designed at A.E.R.E. The circuit employs cold-cathode tubes operating a meter with a full-scale reading equivalent to 350 counts/sec. The meter scale is roughly logarithmic, allowing measurements over a range 0.1–10 mr/hr.

Permissible Activity Levels

The Advisory Panel of the British Medical Research Council have agreed (*1*) that the chief radioactive hazard from drinking water arises mainly from the strontium and barium isotopes present. Chamberlain (*2*) has estimated the emergency permissible ingestion tolerance for strontium and barium in the few days after burst as 30 μc Sr^{89} + 150 μc Ba^{140}, assuming that a man may drink up to 2½ liters of water per day for a 10-day period before an alternative and uncontaminated supply can be found. After discussing the probable extent of leaching out of other fission products during passage through the ground, Chamberlain has established a series of permitted total beta activities (Table 1). These maximum permissible activities have also been agreed to by the Advisory Panel of the Medical Research Council.

The figures in Table 1 are based on an assumed ratio of Sr^{89} to total activity at different times after burst that may be modified at a later date. The reduction in permitted activity at longer times after burst is due to the decreasing ratio of total activity to strontium with time. In addition, the water will have a gamma activity due to the release of 0.7 gamma photon per beta disintegration.

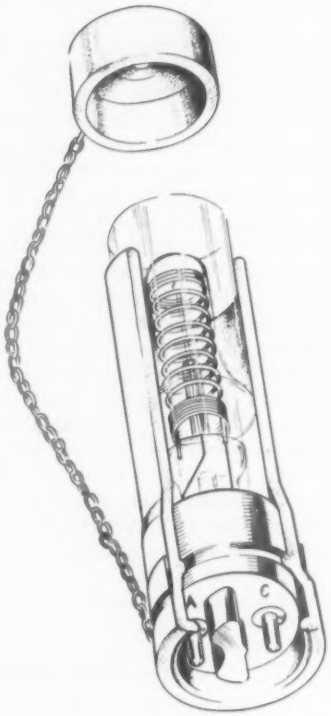

FIG. 1. Liquid beta counter (type CV 2286)

TABLE 1—British Emergency Permissible Tolerance Levels for Drinking Water

Time after burst	*Emergency permissible total beta activity (μc/ml)*
12 hours	1.8×10^{-1}
1 day	6×10^{-2}
2 days	2.4×10^{-2}
10 days	1×10^{-2}

General Method

It was required that water activities corresponding to less than 0.1 tolerance at 10 days (less than 10^{-3} μc/ml) should easily be measured and that the volume of the sample be kept to a minimum; the sensitivity of the counter should enable measurements to be made in the presence of general gamma contamination. For these reasons, the counter had to be sensitive to the fission-product beta radiation.

Three types of counter could, in principle, have been used:

1. A thin-walled "dipping" beta counter operating at a low voltage.
2. An end-window form of the gamma counters that were designed for the contamination meter.
3. A robust form of a "skirt" liquid counter (*3*) operating at a low voltage.

The first two methods would require that the liquid source be presented to the counter in an accurately reproducible manner, and some form of jig mounting would be necessary. In addition, the dipping counter would require a removable shield to protect the exposed thin-wall section when not in use, while the end-window counter would necessitate a thin window, either of mica or a sucked-in glass bubble, which must be of uniform thickness and which would need pro-

tection. The advantage of the latter method would be that many components and techniques employed in manufacture of the existing gamma counters could be readily adapted.

In the third method, all problems of geometry would be overcome since the liquid sample is poured into an annulus surrounding the thin-wall section; the thin wall is protected by a stout outer glass skirt. The neoprene shock-proof jacket designed for the gamma counters could be utilized to protect this skirt. The disadvantage in manufacture would be that a considerable amount of skilled glasswork would be required, though this might be overcome by automatic equipment.

A reduction in counting rate could be obtained for the dipping and end-window counters, should this be necessary when measuring highly active samples, by increasing the distance between counter and liquid surface, or by the use of limiting apertures. However, this would add to the equipment that would have to be carried in the equipment haversack. If a "liquid" counter were employed, this counting-rate reduction could only be done by dilution of the sample, though it would make the method more adaptable to changes in the tolerance levels.

From these considerations, the liquid-annulus type of counter offers the best possibilities.

Construction of Counter

The counter (Fig. 1) was developed in conjunction with Messrs. 20th Century Electronics Ltd., West Norwood, London, England, who had much experience in the manufacture of a high-voltage counter of this type (*3*).

The sensitive volume is 20 mm long × 9 mm diameter. The thin-glass wall is drawn mechanically, and it seems likely that the thickness will be maintained at 13.5–17 mg/cm² over a length of at least 2 cm.

The annular liquid space was kept as wide as possible without exceeding the maximum outside diameter of the gamma counters, so that the liquid would be of "infinite" thickness for fast beta particles, which in turn would enable normal glass tolerances to be used for the outer skirt without large variations in counter sensitivity. The dead-space above the thin-wall section was increased for the same reason.

The liquid capacity, measured to the brim, can be maintained in the range

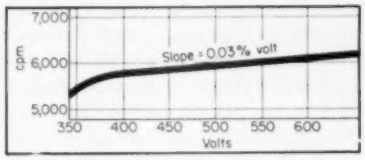

FIG. 2. Type CV 2286 counter plateau

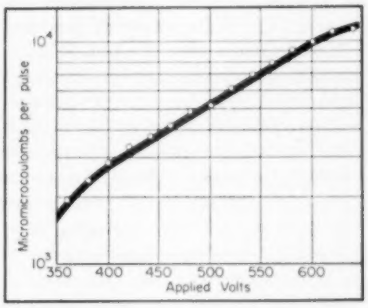

FIG. 3. Charge separated per pulse (type CV 2286)

6.0–8.0 ml; the effect of this variation on counter sensitivity will be discussed in the next section. The counters are filled with neon at 200 mm Hg pressure, to which is added a trace of argon and bromine; the cathode is a wire spiral of a 21% chromium-iron alloy.

The center wire, of 0.7-mm-diameter tungsten, is supported at one end only in a pinch of borosilicate glass, and the counter base is moulded from a nylon-loaded Bakelite, resulting in a minimum resistance between pins of 10^{10} ohms. A water-proof seal is provided by the outer neoprene jacket and the counter has passed the full Service climatic tests.

Characteristics

The G-M threshold voltage is in the range 300–370 volts, and, in the present state of manufacture, the plateau of about half the total production are over 200 volts long, with slopes less than 0.1% volt. The remainder are less good; this was to be expected in view of the dimensions of the counter and the small amount of bromine permissible to maintain the low operation voltage.

As with other types of halogen-quenched counters, the characteristics may be greatly improved if a "stopper" resistor of about 470 kΩ is connected directly to the anode, and the specification is then easily fulfilled. The resistor is included in the base of the counter, where preliminary tests have shown it to be necessary, and the counter is then marked with a yellow "splash." It is hoped to dispense with the resistor as production proceeds. Figure 2 shows a typical G-M plateau curve, and Fig. 3 shows the charge separated by the counter at each counting event as a function of applied voltage, with fixed counting rate. The counter current corresponding to a full-scale meter reading is about 1.2 μa at the normal operating point; the saturation current varies from about 10 μa at the normal operating voltage to 40 μa at 200 volts above threshold.

Beta Efficiency

Initial measurements of beta sensitivity were carried out on a batch of sixteen counters; the test source of $Sr^{89} + Pr^{144}$ (discussed in Appendix) had an activity of 1.14×10^{-2} μc/ml. The net counting rate due to this activity when the counters were filled to the brim, allowing for a background measured with the liquid space filled with distilled water, varied from 4,700–5,900/min, the average being 5,300/min; the spread is expected to be less than ±15% in large-scale production. A variation in liquid level to 2 mm down from the brim made no measurable difference to the count rate.

The counting rate corresponds to a meter deflection of about one-half full scale for an activity at 1 day of one-fourth tolerance, and indicates that dilution is required in the early period after burst if activities above tolerance are to be measured. In fact, ten tolerances at 1 day corresponds to a meter reading of 8 mr/hr at a dilution of ×10, while 0.3 tolerance at 10 days reads 0.1 mr/hr. The reading for any other fraction of tolerance or dilution at the same times after burst may be obtained by simple interpolation.

Energy dependence. As the average beta energy of solid fission products decreases with increasing time after burst (Fig. 4), the same batch of counters was examined with a solution containing Tl^{204}. The maximum beta energy of 0.775 Mev is rather less than the estimated average maximum energy of 10-day solid fission products (0.9 Mev), and, although the beta spectra do not agree in shape, the measurement would indicate roughly the degree of spread to be expected. The average maximum beta energy of fission products in water may not de-

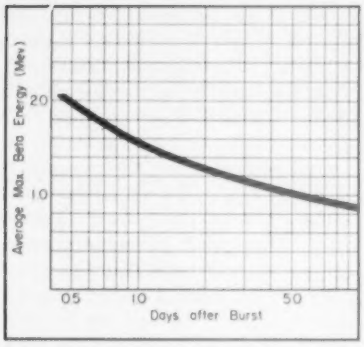

FIG. 4. Average maximum beta energy for solid fission product

crease to the same extent as that of the gross solid products so the result is the worst likely to be encountered.

For an activity of 1.95×10^{-2} μc/ml, the net counting rate varied from 1,700–2,200/min, having a mean value of 1,950/min. The sensitivity spread in production at this energy should therefore be less than ±20%. These tolerances are considerably less than for counters currently employed in liquid assay when used under the same conditions and should, for many purposes, eliminate the need for provision of calibration sources with each instrument. A linear relationship between counting rate and solution activity was found to hold at least up to 2.5×10^4 cps, which corresponds to greater than full-scale reading on the instrument.

The variation of counter efficiency with beta energy must be an important correction in estimating water activity at different times after burst. For this reason, a more comprehensive study was made on one counter (Fig. 5). The efficiency was calculated with the counter brim-full of liquid as this will probably be the easiest method of estimation in practice. For beta-gamma emitters, a correction was made for the gamma sensitivity of the counter by sliding a close-fitting shield of appropriate thickness down over the thin-wall section and embedding it in a thin layer of grease melted into the bottom of the liquid space.

The measured efficiency was only about one-third that of CV 2145 (*3*), due to the wide annular gap and large dead space. However, the counter may be used for the assay of S^{35} in solution, although the counting efficiency measured under the same conditions as above is only 3×10^{-3}%, and may vary by a factor of 2–3 for different counters. The deviation from linearity in Fig. 5, at beta energies below about 1.5 Mev, is probably due to the fact that below this energy the liquid has an effectively infinite thickness, but the curve was not examined theoretically.

The $Sr^{89} + Pr^{144}$ mixture was also measured and the value of efficiency marked on the curve, where it corresponded to a maximum beta energy of 1.65 Mev. This is close to the estimated value for 1-day-old fission products, and although some components were neglected in building the spectrum on which the choice of mixture was made it allows some confidence to be placed in the mixture's correctness.

If it can be assumed that a single isotope of appropriate end point can be used to simulate fission products at other times after burst, then at 4 days a more effective substitute for gross solid fission products would appear from Fig. 5 to be 4.98-day RaE (see Appendix). This has a maximum beta energy of 1.17 Mev, compared with an estimated beta energy for gross solid fission products at 4 days of 1.11 Mev. On account of this variation in counter efficiency, the water activity in μc/ml needed to produce a given meter reading at 10 days (1.1 Mev) is nearly 2.7 times greater than that at 1 day (1.6 Mev).

Gamma Efficiency

The net gamma sensitivity of the counter under the same conditions of measurement was 0.08% at 0.411 Mev, 0.15% at 1.2 Mev, 0.5% at 2.76 Mev. These figures were obtained from the beta-gamma emitters used in obtaining Fig. 4, after correction for the difference in liquid volume produced by the absorber, and the accuracy is probably not better than ±20%. Thus, assuming the average fission-product gamma energy at 1 day to be as high as 1.2 Mev, the counting rate due to gamma radiation will be 3.5% of that due to beta particles. This will increase to 11% at 10 days if the average energies are assumed to be 1.1 Mev (betas) and 0.7 Mev (gammas).

TABLE 2—Liquid Counter Background Due to External Radium Source*

Dose rate (mr/hr)	*Counting rate of CV 2286 counter (cpm)*	*Counting rate of CV 2246, CV 2247 gamma counters (cpm)*
0.2	0.12×10^3	0.42×10^3
0.5	0.3×10^3	1.05×10^3
1.0	0.65×10^3	2.1×10^3
2.0	1.2×10^3	4.2×10^3
5.0	3.0×10^3	1.05×10^4
10.0	6.0×10^3	2.1×10^4

* The figures quoted are the mean of a batch of twelve counters.

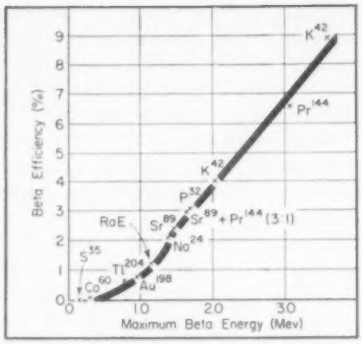

FIG. 5. Beta efficiency of CV 2286, No. HH15, as function of maximum beta energy

The background counting rate, measured unshielded and with the liquid space filled with distilled water, was 10 cpm.

The counting rates due to an external radium source are given in Table 2 and are expressed in terms of the dose rate in mr/hr at the counter due to the source. The liquid space was filled with distilled water for these measurements.

Interpretation of Readings

The instrument scale is calibrated in mr/hr for radium gamma radiation when the gamma counter is mounted at a fixed distance from a contaminated area. Hence, the meter deflections obtained with the liquid counter bear no direct relation to the water activity being measured. However, it should be noted that the indicated reading in mr/hr varies linearly with true counting rate.

Several factors had to be considered before deciding on a method of interpreting the meter readings:

1. The dilution necessary to produce a convenient deflection.
2. The time after burst (see Table 1).
3. The variation in sensitivity with beta energy, which is also dependent on time after burst.
4. Variation in gamma sensitivity with time.

5. The operational requirement that the measured activity should be shown both as a fraction of the tolerance dose and as a concentration in $\mu c/ml$.

A simple circular calculator was constructed, having three concentric discs and a cursor (Fig. 6). All the correction factors listed have been incorporated, and the data on which the calculator is based are given in Fig. 7. The scales are logarithmic to give the same type of "meter-reading" scale as exists on the contamination meter.

Instructions are carried on the reverse side of the calculator. As illustrated, the calculator is operated by setting the dilution factor to the appropriate time after burst on the outer disc, while the time after burst on the inner disc is set to the arrow T on the intermediate disc. If the cursor is now moved to the indicated meter reading, the tolerance level represented by the undiluted sample and the corresponding activity in $\mu c/cm^3$ may be read off under the cursor on the two inner discs. Time after burst has to be fed in twice since two different empirical corrections have to be made, one to both activity and tolerance scales (beta + gamma counter efficiency) and the other to the tolerance scale only (Table 1).

Calibration

To calibrate counters, a jig incorporating a radium-impregnated sintered silver source has been designed (Fig. 8). The foil, developed at the Radio-Chemical Center, Amersham, England, has a "front" thickness of 0.001 in., producing little beta attenuation. The total activity of a foil of area 36 cm^2 is 0.02 μc, which produces a meter deflection up to the calibration mark for an "average" counter when the instrument calibration control is in the middle of its range.

Calibration procedure is to insert the counter into the jig tube and slide down the foil by means of a collar to which it is attached until it touches the bottom of the counter annular space. The foil expands tightly against the inside of the glass "skirt," when the calibration control is adjusted to give the correct deflection.

On the batch of counters discussed in the section on beta efficiency, the spread in counting rate with a source of this type was $\pm 12\%$. The readings from each counter could be accurately correlated with readings for the same counter with the liquid source, so that the jig can be regarded as a satisfactory substitute for a liquid standard. These foils can be produced with a tolerance on activity of $\pm 5\%$.

Errors

The causes of error in estimating the tolerance level and activity in $\mu c/ml$ of a particular water sample may be divided into two groups.

Instrumental errors. The results indicate that the variation in counting rate of different counters, when measuring identical samples, should never be greater than $\pm 20\%$, and will probably be less than this figure. If the instruments are initially set up with calibration sources, the maximum possible error at the calibration point between any two instruments, if the counters are changed without further calibration, will therefore be $\pm 50\%$, including counter and calibration tolerances but excluding reading errors. Due to component tolerances in the instrument, the maximum error anywhere else on the scale may be $\pm 60\%$ on some instruments. If the setting-up procedure is carried out whenever a counter is changed, then the maximum error at any point on any scale will fall to about $\pm 25\%$.

Conversion errors. When the instrument reading is converted into a tolerance level or an activity in $\mu c/ml$, a more important source of error, and one that is almost impossible to assess, will be the uncertain composition of the fission-product activity in water. The extent of leaching out will depend markedly on the nature and stratification of the ground on which the water either falls or through which it passes prior to assay; this may modify the beta spectrum. In view of the variation in counter efficiency with average maximum beta energy (Fig. 3), this would produce errors in the estimated activity.

The conversion to a fraction of the

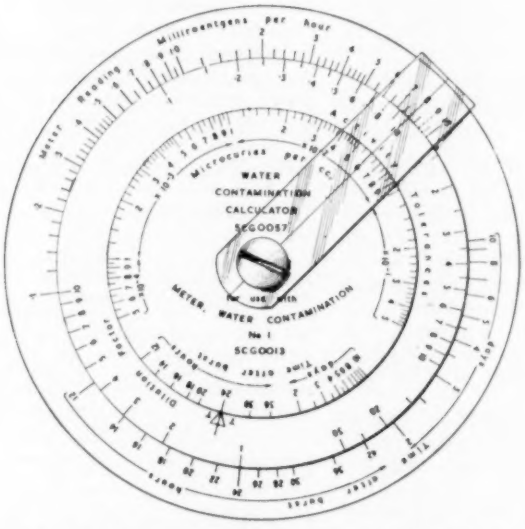

FIG. 6. Water contamination calculator. Tolerance level and corresponding activity can be determined at desired time after burst

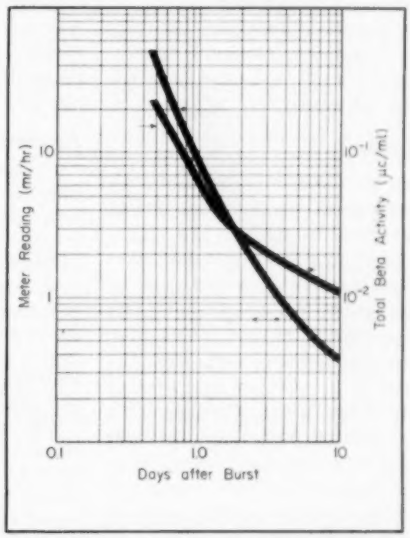

FIG. 7. Meter reading and total water activity (beta) to give 1 tolerance as a function of time

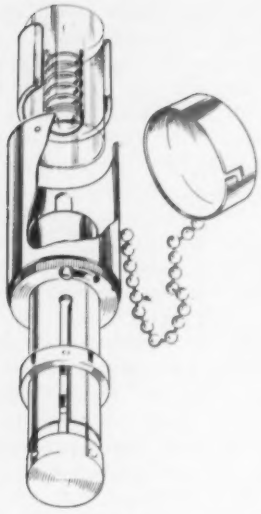

FIG. 8. Calibration source

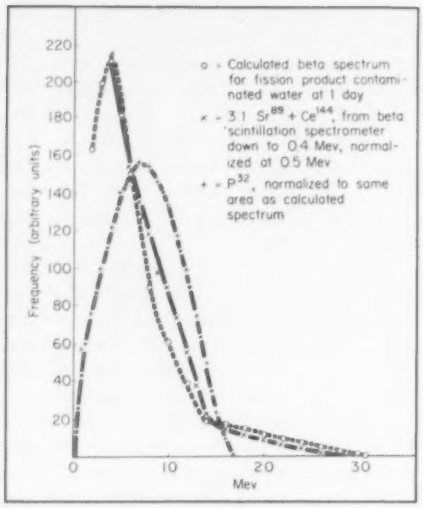
o = Calculated beta spectrum for fission product contaminated water at 1 day
× = 3.1 Sr^{89} + Ce^{144}, from beta scintillation spectrometer down to 0.4 Mev, normalized at 0.5 Mev
+ = P^{32}, normalized to same area as calculated spectrum
Frequency (arbitrary units)
Mev

FIG. 9. Fission-product beta spectra

tolerance level is subject to an additional uncertainty since, if the ratio of Sr^{89} to total activity differs appreciably from that assumed in fixing the tolerance levels (*2*), the true tolerance level of a water sample will not be the same as that indicated even if all other sources of error could be eliminated.

Thus, an assay carried out in terms of water activity in μc/ml will probably be considerably more accurate than if the tolerance scale is used; an error in the form of this scale on the calculator will not affect the activity readings, for in this sense the two parameters are independent.

These uncertainties will apply to any method that does not measure directly the Sr-Ba activity and must limit the reliability of the survey.

Operation

When the counter is operated with the instrument, the following points must be borne in mind:

1. The counter has been made waterproof and has been protected as far as possible against mechanical shock, but it will inevitably be less robust than the gamma counters used with the instrument.

2. Any liquid with a specific gravity not far removed from 1 may be used as a diluent, so long as it is not likely to be contaminated. Thus, most aqueous liquids stored in bottles, casks, or tanks will be satisfactory.

3. No special apparatus need be supplied for dilution; the counter itself will suffice as a measuring cylinder, so long as another clean container is available for mixing.

4. Before making a measurement, the background should be recorded with the counter filled with clean liquid. If this reading is appreciable due to contamination of the surroundings, it must be subtracted from the reading obtained with the active liquid before conversion to μc/ml or tolerance.

5. If this background reading is made after dilution, then any beta contamination of the counter may also be corrected for.

6. Repeated washes with clean water will reduce beta contamination of the counter, but if this is serious it will be necessary to return the counter for specialized treatment.

Appendix

Phosphorus-32 has been suggested as a suitable test source since its maximum beta energy, 1.69 Mev, is close to the average maximum energy for 1-day-old fission products, 1.6 Mev (Fig. 3). However, a plot of the 4-day fission-product spectrum, together with that for P^{32} (*4*), shows gross differences between the two spectra, since P^{32} contains a much higher proportion of beta particles in the high-energy range.

The table of fission yields compiled by Hunter and Ballou (*5*) were used for the present study, but only those components expected to be present in drinking water were included. All decay chains originating in noble gases, the rare earths, and zirconium were excluded (*3*). This left the strontium, barium, lanthanum and iodine isotopes, together with several small contributions. It is probable that iodine will be present only as a fraction of its initial concentration, but, as this is not certain, it was included to the full extent.

The composite spectrum for 1 day was built up by assuming a triangular frequency-energy spectrum with the apex at 0.31 of the maximum energy, the area being proportional to the fission yield. The general shape of the spectrum was not very sensitive to changes in estimated constitution because of the large number of components. The resulting curve was roughly analyzed into two components. A mixture of Sr^{89} + (Ce^{144} → Pr^{144}), in the ratio of activities 3:1:: Sr^{89}:Pr^{144}, gave a satisfactory fit. A thin source having these proportions was made up and analyzed with a beta scintillation spectrometer (*6*), down to 0.4 Mev. The results, together with P^{32} (*7*), all normalized to the same area, appear as Fig. 9.

Phosphorus-32 fits the 1-day fission spectrum rather more closely than that at 4 days, as given by Hursh (*4*), but the Sr^{89}–Pr^{144} mixture is appreciably better, at least down to the lowest energy plotted. Exact agreement at very low energies is not as important in this application, since the counting efficiency of any G-M counter for an external source falls off rapidly in this region. At times later than 1 day, the suggested mixture will fit progressively less well, and a different recipe may be necessary unless other methods are available.

The over-all efficiency for P^{32} lies very close to that of the Sr – Pr solution (Fig. 4), and hence, on the basis of this measurement, it must be concluded that P^{32} is a satisfactory substitute in this counter for the fission products likely to be the main contaminants in drinking water at 1 day. The result may be explained by the fact that, although the P^{32} spectrum has an excess of beta particles in the energy range 0.8–1.4 Mev, there is of necessity an absence of betas above 1.7 Mev, the effects of which would oppose each other in a counter.

* * *

The author wishes to thank Miss N. Wildblood who prepared the radioisotopes and Mr. R. B. Owen, who measured the Sr^{89}–Pr^{144} beta spectrum. This article is published by permission of the Director, Atomic Energy Research Establishment, Harwell. *Crown copyright reserved.*

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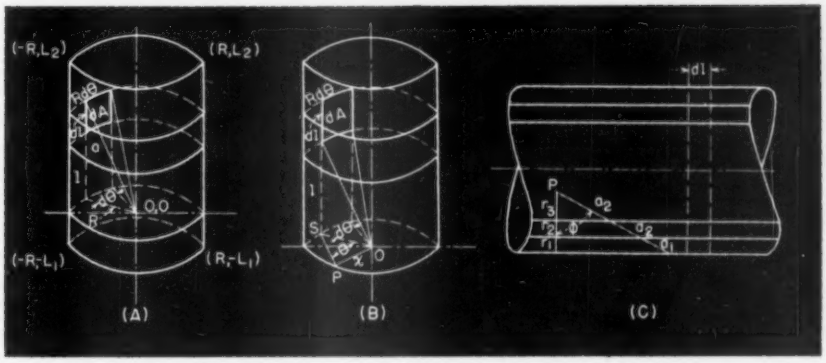

FIG. 1. Parameters for calculation of flux distribution in cylindrical source

Measurements of Gamma-Ray Dosage in Finite Absorbers

In the utilization of γ-ray sources for sterilization, the problem of dosimetry arises. Measurements of a cylindrical Co^{60} source and concentric absorbers with polyvinyl chloride dosimeters agree well with calculated dosage distribution

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IN THE DESIGN of a radiation-processing installation, three factors must be determined by dosimetry: attenuation of the photon beam and dosage distribution and energy absorption in the irradiated sample. The approach described here combines calculations based on Beer's Law with polyvinyl-chloride-dosimeter measurements.

Attenuation of the Photon Beam

Attenuation of the photon beam can be calculated only under very special conditions of equilibrium. Absorption coefficients, which are in theory functions only of the wavelength of incident radiation and atomic number of the absorber, become in practice functions of the beam and absorber geometry.

For the case of cobalt gamma rays, two equilibrium conditions may apply: (1) secondary electron equilibrium, where an additional slice of absorber adds as many electrons as it absorbs, and (2) secondary photon equilibrium, where an additional absorber generates as many photons as are absorbed. Under conditions of secondary electron equilibrium, where Compton absorption is the chief process, the Compton absorption coefficient, σ_a, describes the beam attenuation; under secondary photon equilibrium, the total Compton coefficient, $\sigma_a + \sigma_s$, describes the beam attentuation.

Dosage Distribution in the Sample

Design of sterilization processes must in general be based on the point in the medium that receives the lowest dosage. Chemical and physical dosimeters developed thus far are not convenient for making these measurements directly, particularly in small-volume elements.

Energy Absorption in the Sample

From the standpoint of chemical and biological effects, or dose measurements, it is the energy, and therefore the gradient of the flux absorbed in the sample rather than the flux itself, that need be considered.

The intensity gradient can be related directly to the dose by conventional dosimeters only under the two conditions stated: (1) under secondary electron equilibrium, where the Gray-Bragg principle applies, and (2) under secondary photon equilibrium when $(I/I_0)/(D/D_0) = 1$, where D = dose and I = intensity.

There are two ways of answering these three problems:

1. Experimentally by means of a very small dosimeter, such as the polyvinyl chloride films. The dosage distribution and the energy absorbed can then be measured directly, and the beam attenuation may be calculated.

2. Determine the attenuation of the beam by assuming a Beer's Law absorption and allow for the geometry factors. From this one may calculate the dosage and the dosage distribution. However, the Beer's Law coefficient must be correctly chosen.

A combination of the two techniques

can therefore be used to establish apparent absorption coefficients. The measurements and the calculations are applicable only to the specific system under consideration, but the principles and the general approach apply to any problem in dosimetry.

Experimental Procedure

The polyvinyl chloride dosimeter used was developed by Henley and Miller (*1*). The Co^{60} radiation source is a prototype of the one described by Manowitz (*2*). It consists of a cobalt pipe, 13.5-in. long, 2.8-cm o.d., that has been activated in the Brookhaven Reactor. The pipe is encased in aluminum and enclosed in a lead vault.

The irradiations were carried out by placing the samples in an aluminum canister, which is then lowered by remote control into the vault and centered so that the canister is concentric with, and inside of, the Co^{60}. The dose rate in air was 72,000 r/hr at the center of the source.

Polyvinyl chloride films, ¾ × ½ in. and 0.0016 in. thick, were placed between "sandwiches" of the various irradiated materials. Rods or pipe of Lucite, aluminum, iron, copper, and lead were fabricated into concentric cylindrical absorbers and telescoped to fit into the aluminum basket. Except for the aluminum, all metals used were more than 99% pure. The aluminum 61 ST alloy contained 1.5% Cu, 0.7% Fe, 0.2% Zn, and 0.15% Cr. The absorbers were machined to fit inside of each other with a clearance of 0.004 in. or less.

Discussion of Results

Insofar as the results may be used to obtain answers to the questions of attenuation, dosage distribution, and energy absorption, it may be useful to discuss the data in the light of these concepts.

Attenuation of the Photon Beam

Unless the incident energy is determined absolutely by an absorption calorimeter, this measurement must be made under equilibrium conditions with ion chambers or with film or chemical dosimeters. The polyvinyl chloride films were calibrated by placing them in the same spot, under the same conditions, as an ion chamber that recorded D roentgens (D and I in this discussion are per unit time over one time interval); then D was set equal to the color change in the films.

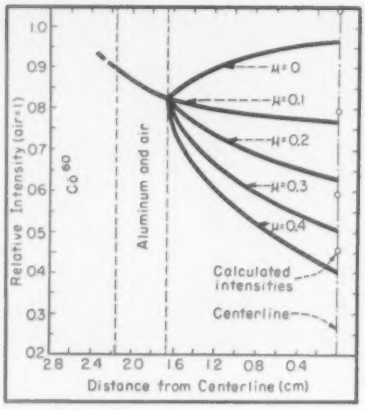

FIG. 2. Calculated variation of flux intensity with distance from source axis

With the films placed in two air cavities in a solid absorber along the axis of a photon beam, x cm apart, assume that one film records a color change equivalent to D, and the other a change equal to D'. If the equilibrium conditions are identical to those under which the films are calibrated, these two measurements give the photon flux at the two points. Since the film was calibrated in roentgens, some primary conversions must be made to calculate the photon flux.

The incident energy can be calculated from a formula derivable directly from the definition of the roentgen as a number of ionizations per gram of air

$$\frac{5.18 \times 10^{-11}\text{erg}}{\text{ionization}} \times 2.08 \times 10^{9} \frac{\text{ionization}}{\text{cm}^3\ \text{air}} = 0.108\ \text{ergs cm}^{-3}$$

Then, if I is the incident energy in ergs/cm² and μ is the absorption coefficient of the air at that wavelength

$$\mu I = 0.108\ D$$

for Co^{60} γ's, taking

$$\mu = 3.5 \times 10^{-5}\ \text{cm}^{-1}$$
$$I = 3.09 \times 10^{3}\ D\ \text{erg/cm}^2$$

This is the flux incident on both the films and the ion chamber when they read D roentgens.

Another method of calculating the photon beam (I) is to measure the flux at one point (I_0), and then, assuming a Beer's Law absorption and inverse-square effects

$$I = I_0\, e^{-\mu x}/x^2$$

For a cylindrical source, there are three variables that affect the flux distribution inside a source of finite length:

Position of sample with respect to vertical axis. Consider the intensity at (0,0) due to the radioactive element dA (Fig. 1A). In cylindrical coordinates

$$I_0 = K\alpha,\ I = \frac{I_0 e^{-\mu a}}{a^2},\ dA = Rd\theta dl$$
$$\alpha = N/A,\ d\alpha = -(N/A^2)\, dA$$

where I_0 = intensity due to a unit amount of activity at a unit distance in r/hr, α = curies per surface area, a = distance from active element to a point, K = experimental constant, and μ = absorption coefficient.

$$dI = -\frac{KN}{A^2}\frac{e^{-\mu a}}{a^2}\, dA$$

Solving

$$I = -\frac{2I_0 R\pi}{A}\int_{-L_1}^{L_2}\frac{e^{-\mu a}}{a^2}\, dl \qquad (1)$$

In spherical coordinates

$$I = -\frac{4I_0}{A}\int_0^{\frac{\pi}{2}}\int_{\tan^{-1}\frac{L_1}{R}}^{\tan^{-1}\frac{L_2}{R}} e^{-\mu R\sec\phi} d\phi d\theta \qquad (2)$$

Equation 2 has been solved by Sievert (*3*), and the values are tabulated by Wilson (*4*).

Position of sample with respect to horizontal axis. Contributions to a point off the axis (Fig. 1B) are given by

$$\overline{a^2} = l^2 + \overline{ps}^2$$
$$\overline{ps}^2 = x^2 + R^2 - 2Rx\cos\theta$$
$$a^2 = x^2 + R^2 + l^2 - 2Rx\cos\theta \qquad (3)$$

Substituting in Eq. 1

$$I = -\frac{2I_0 R\pi}{A} \times \int_{-L_1}^{L_2}\frac{e^{-\mu(x^2+l^2+R^2-2Rx\cos\theta)^{\frac{1}{2}}}}{x^2+l^2+R^2-2Rx\cos\theta}\, dl \qquad (4)$$

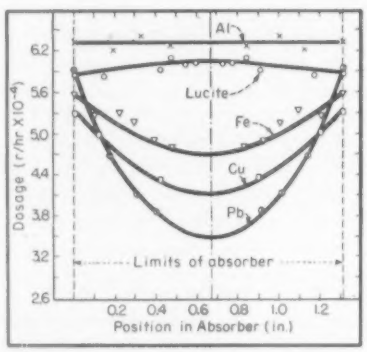

FIG. 3. Measured dosage distribution

Equation 4 holds for the simple case where the cavity is filled with a uniformly absorbing, homogeneous medium. The integral is identical with the one given by Sheppard (*5*).

Effect of nonhomogeneous absorbers. When the source cavity is filled with three concentric absorbing cylinders, *a*, *b*, and *c* in thickness, the mathematics become more involved. The rigorous solution is unmanageable, and due to the complete lack of symmetry, there appears to be no easy simplification that would permit a graphical integration.

Since the gamma rays travel through at least 0.6 cm of cobalt and two layers of aluminum before they reach the sample, the case of nonhomogeneous absorption is important enough to warrant an approximate solution.

Assuming a cosine relationship (Fig. 1C)

$$a_1 + a_2 + a_3 = a$$

$$\cos\phi = \frac{r_1 + r_2 + r_3}{a} = \frac{r_1}{a_1} = \frac{r_2}{a_2} = \frac{r_3}{a_3}$$

$$I = \frac{-2I_0R\pi}{A}\int_0^{2\pi} \times \int_{-L_1}^{L_2} \frac{e^{-(\mu_1 a_1 + \mu_2 a_2 + \mu_3 a_3)}}{a^2}\, dl d\theta$$

$$= \frac{-2I_0R\pi}{A}\int_0^{2\pi}\int_{-L_1}^{L_2} \frac{e^{-\frac{(l^2+R^2+x^2-2Rx\cos\theta)^{\frac{1}{2}}}{r_1+r_2+r_3}(\mu_1 r_1 + \mu_2 r_2 + \mu_3 r_3)}}{l^2 + R^2 + x^2 - 2Rx\cos\theta} \times d\theta dl \quad (5)$$

Equation 5 has been solved numerically by methods of mechanical cubature, and the results are presented in Fig. 2.

Fortunately the assumptions made constitute a rather good approximation; at the center, where the solution becomes exact, a comparison between Eqs. 4 and 5 gives the expected results. The intensities predicted by Eq. 4 are high for the low μ values and low for the high μ values. In Fig. 2 this is shown by the circles that were calculated from Eq. 4.

Dosage Distribution in the Sample

Figure 3 is a plot of the experimental data obtained with the polyvinyl chloride films. The general shape and characteristics of these curves compare quite well with the calculated curves of Fig. 2. An interesting result is the very high intensity at the surface of the lead absorber. This can be ascribed to the high back-scattering of secondary electrons from the lead cylinder (*6*).

Comparison of Calculated and Experimental Absorption Coefficients

	Intensity at center					
	Measured		*I*	$\sigma_a + \tau$ (cm^{-1})		
Absorber	(*ergs/cm²/hr* × 10^{-7})	*Relative to air*	(*ergs/cm²/hr* × 10^{-7})	*Calculated*	*Experimental*	*E*
Air	22.3	1	—	—	—	—
Lucite	19.8	0.89	19.8	0.032	0.031	6.3
Al	18.8	0.84	18.5	0.07	0.065	12.1
Fe	14.5	0.65	15.5	0.19	0.185	28.8
Cu	12.5	0.57	14.0	0.22	0.235	32.8
Pb	10.5	0.47	13.1	0.37	0.34	43.5

The sets of data for iron and copper were analyzed statistically to determine if the difference of 40 between the sets is significant. The standard deviation of the difference is 1; therefore the 95% confidence limit is 40 ± 2.2, and there can be no question about the significance of the difference.

Total Dosage Absorbed by Sample

The dosage absorbed can be directly determined by averaging the experimental values obtained by the films. It can also be obtained from the intensity calculations in an indirect manner.

If a plot of I versus dosage such as Fig. 2 is calculated at a series of points x cm apart, the energy absorbed in ergs/cm³, or roentgens, will be

$$D = \bar{I}\mu = \frac{\mu\int_{x_1}^{x_2} I dx}{x_2 - x_1} \quad (6)$$

where $\bar{I}$ = average photon intensity.

A method of determining D is therefore: (1) calculate the theoretical intensity in terms of the inverse square law and a Beer's Law absorption, and (2) determine the apparent absorption coefficient under the conditions of the experiment.

The apparent absorption coefficients were obtained by matching the intensity (relative to air) at the center of the source as calculated from the polyvinyl chloride film measurements with the corresponding intensity from Fig. 2, which was calculated using an arbitrary μ. These experimental coefficients are given in the table. The calculated $\sigma_a + \tau$ was obtained by averaging the values of White (*7*) and Davisson (*8*). There is a good agreement between the theoretical and experimental data. This is somewhat surprising for the heavier elements, since, if a photon path length of about 3.5 cm is assumed, it would be expected that a considerable amount of the scattered radiation has been absorbed. Since there are no other data available, it is a difficult matter to judge where and if a discrepancy exists.

The energy absorbed by the sample is given in the table. It was obtained by multiplying the average intensity (obtained by numerical integration of Eq. 6) by the experimental absorption coefficient. This procedure is not absolutely reliable since it is likely that the absorption coefficient is not constant throughout the sample.

This cannot be easily remedied. It is, for all practical purposes, impossible to solve the exact field geometry equations. The approximate solution to the field geometry problem, Eq. 5, is only rigorous near the center of the source.

For any design or shielding calculations, it would be convenient to obtain the absorption coefficients as a function of distance from the beam

$$\mu = \tau + \sigma a f_1(x/x_a) + \sigma_s f_2(x/x_s) \quad (7)$$

where x_a and x_s are the distances for secondary electron and photon equilibrium.

It will require a broad experimental program to determine the functions f_1 and f_2, and the quantities x_a and x_s.

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Radiation Decomposition of Carbon-14-Labeled Compounds

Self-irradiation produces much more decomposition than had been expected for several labeled organic compounds. Data to date are reported here, and methods for determining radiopurity and preventing decomposition are considered

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RECENT OBSERVATIONS at several laboratories (*1–3*) have shown that organic compounds labeled with carbon-14 are capable of extensive decomposition under the influence of their own radiation. In view of the volume of research carried on with the aid of these compounds, it is important that researchers recognize this hazard to correct interpretation of their experimental results.

The previous confidence felt by researchers that there was little probability of serious decomposition was held largely because of the low average energy (49–50 kev) of the beta particles emitted by the isotope.

Although nearly all the betas are absorbed in a solid sample of a labeled compound, and the specific ionization is thus very high, only a small amount of radiation decomposition should be expected. For example, in the case of a compound containing one millicurie of C^{14} per millimole (a rather high specific activity), it has been calculated (*2*) that the radiation should destroy the compound at a rate of only 0.3% per year. However, the data presented in this article suggest that occasionally decomposition may proceed through some type of chain reaction, and that destruction of the labeled compound may thus be far more severe than expected. It is the possibility of this enhanced decomposition that is particularly disturbing.

Examples of Self-Decomposition

The data gathered so far on the radiation decomposition of C^{14}-labeled compounds are summarized in the table on the next page.

Calcium glycolates. Decomposition was observed by S. Weinhouse (*1*). He found that both glycolates decomposed to give radioactive formate.

Since several thousand molecules are decomposed by each beta particle emitted, there seems no reason to expect that the amount of radiation damage should be dependent upon the particular source of the C^{14} betas, i.e., whether the glycolate is 1-labeled or 2-labeled. The difference between the G values may have arisen from the production of volatile radioactive decomposition products that escaped detection. It is therefore possible that the G values actually are equal.

Methyl-labeled choline chloride. This compound $[(C^{14}H_3)_3NCH_2CH_2OH]^+Cl^-$ is the most striking example of radiation decomposition (*1*). Severe destruction (63%) was observed even though the anhydrous choline chloride samples had been stored in the dark in sealed evacuated tubes. The main products of the decomposition were radioactive trimethylamine and inactive acetaldehyde; thus the principal result was the breaking of the bond between the nitrogen and carbon atoms of the ethanol group.

The abnormal sensitivity of choline chloride to radiation damage has been further demonstrated (unpublished work) at the Radiation Laboratory of the University of California by exposing the unlabeled compound to the γ-radiation of a Co^{60} source and to the β-radiation of a 3-Mev linear electron accelerator. In both experiments the choline chloride was found to be very sensitive to the radiation. The γ-radiation (1.1 Mev, 2.7×10^7 rep) caused 67% decomposition; the β-radiation (5×10^6 rep) caused 51% decomposition. That a given amount of γ-radiation causes less destruction than the same amount of β-radiation is to be expected since, for a given energy, the specific ionization induced by the former is far lower.

Methanol. This compound was prepared by the Oak Ridge group (*3*) with a specific activity (8–10 mc/mmole) that is one of the highest ever reported. After 21 months of storage in sealed ampoules, the methanol was found to have yielded considerable gas (5.4 molecules per 100 ev, identified as methane and hydrogen) along with material less volatile than methanol. These latter products were identified as glycol, glycerol and erythritol ($HOCH_2$—CHOH—CHOH—CH_2OH), and they were present in the ratio of 1,360:15:1. The methanol seems to exhibit a chain reaction in its decomposition, but the chain is far shorter than it is in the case of choline.

Methyl iodide. Wagner and Guinn (*2*) made an accurate spectrophotometric estimation of the free iodine

C^{14}-Labeled Compounds Decomposed by Self-Irradiation

Compound	Amt. radiation (rep × 10^{-6})*	Decomposition (%) Observed	Calc.†	Observed G (molecules decomp./ 100 ev)	Ref.
Calcium glycolate-1-C^{14}	25.4	13	1.3	31	1
Calcium glycolate-2-C^{14}	17.5	22	0.9	77	1
Choline-methyl-C_1^{14}-chloride	10.7	63	0.4	490	1
Methanol-C^{14}	593	19	5.1	12	3
Methyl iodide-C^{14}	3.2	0.14	0.12	3.6	2
Norleucine-3-C^{14}	17.4	2	0.6	10	1
Norvaline-3-C^{14} · HCl	40.3	5	1.7	9	1
Valine-4,4′-C_2^{14} · HCl	8.2	1	0.3	9	1

* The relationships for the calculations of these figures are given in ref. (*1*). Differences between some of these figures and those of (*1*) are due to small errors in the earlier calculations.

† These calculations are made with the assumptions that (a) all of the emitted beta particles are absorbed by the compound itself, (b) the decomposition is caused solely by the ionisation produced by electron impact, (c) the average energy loss is 32.5 electron volts per ion pair formed, (d) the ionization always results in molecular decomposition, and (e) there is no recombination of dissociated radicals or ions to the original molecule.

produced in this labeled compound during its storage in evacuated tubes. This measurement agrees closely with the decomposition calculated on the basis of no chain reactions. It is very probable that the great majority of C^{14}-labeled compounds will decompose at about this rate.

Norleucine, norvaline and valine. These were shown, via paper chromatography and autoradiography (*1*), to show small amounts of decomposition. Through the use of the same methods, comparable amounts of self-radiation have been shown (*1*) to give less than 1% decomposition in the following compounds: glycine-2-C^{14}HCl; guanine hydrochloride-4-C^{14}; 8-azaguanine-4-C^{14}; adenine sulfate-4,6-C_2^{14}; 8-aza-adenine-4,6-C_2^{14}; stilbamidine diisethionate-amidine-C_2^{14}; thyroxine-α-C^{14}; and succinic acid-2-C^{14}.

Determination of Radiopurity

In general, ordinary chemical and physical tests are not adequate to determine the presence of small quantities of radioactive breakdown products. Such labeled impurities may be a negligible fraction of the total material but, at the same time, a large fraction of the total radioactivity.

The most convenient general method for establishing the presence or absence of nonvolatile radioactive impurities is by preparing a "two-dimensional" paper chromatogram of the material to be tested and, finally, preparing an X-ray-film autoradiograph of the chromatogram. This autoradiograph will have one spot for each labeled species in the chromatographed material; in addition, the fraction of radioactivity present in each radioactive impurity may then be determined directly on the paper with a thin-window G-M tube. If only one spot is observed on the X-ray film, a high degree of radiopurity is indicated.

If the presence of a volatile radioactive impurity is suspected, a paper chromatogram may be prepared so that the impurity will not be lost. For example, the volatile trimethylamine that resulted from decomposition of labeled choline chloride was retained on a paper chromatogram through the use of an acidic solvent (*1*). If this procedure cannot be followed, any volatile decomposition products may be removed by dissolving the labeled compound in an appropriate solvent and then evaporating the system to dryness under vacuum. Finally, after possible volatile impurities are thus removed, the paper chromatography may then be employed to determine if any nonvolatile radioactive impurities are present. There are several references in the literature (*5–8*) to such use of paper chromatograms.

Prevention of Decomposition

There are several methods available by which the effects of radiation decomposition may be minimized:

1. Low specific activity. The specific activities of labeled compounds should be kept as low as possible. Such low-level C^{14}-assay instruments as scintillation counters and ionization chambers reduce the need, for most research problems, for labeling to the point where radiation decomposition may become troublesome.

2. Dilute solutions. Storage of a labeled compound in dilute solution will minimize radiation effects by providing an absorber (the solvent itself) for the radiation. Although secondary chemical effects from free radicals formed in the solution may contribute to radiation decomposition, these effects will be extremely small if the solution is dilute and if the solvent is chosen with care. Solvents such as carbon disulfide and benzene, which produce few free radicals when subjected to radiation (*4*), should be excellent.

3. Solid additives. Solid labeled compounds may be stored as an intimate mixture with a large amount of an inert, finely-divided solid, such as powdered glass. In this manner, other material would be present to absorb radiation, and the labeled compound could be separated by solution immediately prior to use. However, for this method to be effective the labeled compound must be reduced to a very fine powder. The maximum range of C^{14} beta radiation (150 kev) is approximately 28 mg/cm² (*9*) and the range of the average particle (50 kev) is only 9–10 mg/cm². This means that in a substance with a density of 1 gm/cm³ the range of the average C^{14} beta radiation is only 0.09–0.10 mm. Therefore, the solid particles of a substance with this density should be reduced to a size where the average diameter is at least less than 0.1 mm.

* * *

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FLUORINE-18: Preparation, Properties, Uses

Fluorine-18 is available by a number of convenient nuclear syntheses, opening up a wide potential for tracer studies. The authors discuss the most practical methods for preparing the nuclide and working with it to study a wide variety of chemical problems

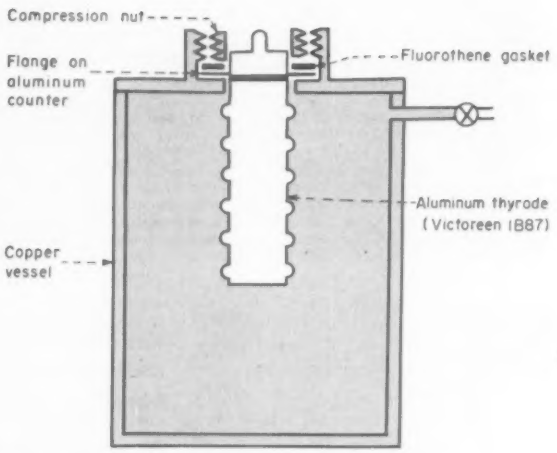

FIG. 1. Copper vessel for gas-phase counting

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THE CHEMISTRY of fluorine has attracted ever increasing attention in recent years. Since 1940, when both inorganic and organic compounds of fluorine began to play an important role in the field of nuclear energy (*1*), interest in this element has steadily mounted. The important work of Emeléus and his colleagues on the halogen fluorides (*2*), the development of industrially important fluorine plastics [polychlorotrifluoroethylene (*3*), polytetrafluoroethylene (*4*)], and successful syntheses of fluorocarbon chemicals by Cady, McBee, Miller, Simons and others, all serve to illustrate the current activity in this burgeoning field. Two recent volumes by Simons (*5*) and by Haszeldine and Sharpe (*6*) summarize much recent work. The biological significance of fluorine as a trace element has also been responsible for much recent research. In considering the future course of research in these and other fields of fluorine chemistry, it becomes appropriate to examine possible contributions that tracer techniques may be able to make.

Preparation of Fluorine-18

Although fluorine, occurring in nature, is monoisotopic, a number of radioisotopes have been synthesized. The longest-lived of these is F^{18}, a positron emitter with a half-life of 112 min. While this half-life is rather short, nevertheless, it is possible to utilize it for a wide variety of studies. This nuclide was first described by Snell (*7*) and by Pool, Cork, and Thornton (*8*) in 1937. Snell observed a 112 ± 4 min activity which he characterized chemically as a fluorine isotope, on bombarding neon gas with 5-Mev deuterons. Pool *et al.* (*8*) obtained a fluorine activity by bombardment of F^{19} with fast neutrons and attributed the formation of F^{18} to an $(n,2n)$ reaction. Both characterized this activity as a positron emitter.

Since then, F^{18} has been detected as a reaction product in a large number of nuclear reactions. These are summarized in Table 1; the order follows that of Hollander, Perlman, and Seaborg's table of isotopes (*9*).

As Table 1 shows, a wide variety of nuclear reactions will yield F^{18}. Not all of these, however, are of practical significance; methods that are of preparative value are discussed later. Here we will point out only that both cyclotron and betatron bombardment can be used effectively, and that the nuclear reactor (*15*) can be used to very good advantage in F^{18} preparation.

Considering the nuclear properties of F^{18}, it was soon observed that F^{18} decays by positron emission. Zah-Wei Ho (*27*) reported that the β^+ spectrum was two-fold complex, and that a gamma ray was associated with decay. Knox (*28*), however, found that no nuclear gamma ray was associated with the decay of F^{18}. Blaser, Boehm, and Marimer (*29*) in turn found that the β^+ spectrum was simple, with an energy of 635 ± 15 kev. Ruby and Richardson (*23*) confirmed this observation and determined an energy of

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TABLE 1—Formation of F^{18} by Various Nuclear Reactions

Nuclear reaction	*Energy of bombarding particle (Mev)*	*Target*	*Yield*	*Physical constants reported*	*Ref.*
$O^{16}(\alpha,pn)F^{18}$ or $O^{16}(\alpha,2n)F^{18}$	40	PbO	Erratic	$t_{1/2}$ = 107,115 ± 10 min	*10*
$O^{18}(p,n)F^{18}$	4	NaOH		$t_{1/2}$ = 107 min β^+ = 0.76 ± .07 Mev	*11*
$O^{17}(d,n)F^{18}$	2	O_2, CdO, H_2O WO_3		$t_{1/2}$ = 112 min β^+ = 0.7 Mev	*12*
	3.45	PbO		$t_{1/2}$ = 114 min β^+ = 0.7 Mev	*13*
	3.7	WO_3		$t_{1/2}$ = 114 min	*14*
$O^{16}(t,n)F^{18}$	2.65	$LiNO_3$	2×10^{-5} F^{18} per t	$t_{1/2}$ = 110 min β^+ = 0.7 Mev γ = 0.56 Mev	*15*
$F^{19}(n,2n)F^{18}$				$t_{1/2}$ = 108 min; β^+	*8*
$F^{19}(d,t)F^{18}$	9	NaF	Cross section 3.9 $\times 10^{-27}$ cm^2	$t_{1/2}$ = 112 ± 2 min	*16, 17*
$F^{19}(\gamma,n)F^{18}$	20	LiF	3.48 mb	$t_{1/2}$ = 112	*18, 22*
	100	NaF, LiF		$t_{1/2}$ = 112	*19*
	50–100	LiF	2.7 rel. to $N^{14}(\gamma,n)N^{13}$	$t_{1/2}$ = 110 β^+ = 0.7 Mev	*20*
	70			$t_{1/2}$ = 112 min	*21*
$F^{19}(p,pn)F^{18}$		AlF_3		$t_{1/2}$ = 112 β^+ = 649 ±9 kev	*23*
$Ne^{20}(d,\alpha)F^{18}$	5	$Ne_{(gas)}$		$t_{1/2}$ = 112 ± 4 min. β^+ max 500 kev	*7*
$Na^{23}(\gamma,\alpha n)F^{18}$	50, 87	Na_2CO_3, $NaNO_3$	0.17 rel. to $N^{14}(\gamma,n)N^{13}$	$t_{1/2}$ = 112 min	*24*
Spallation reactions with high-energy protons	420				
O^{18}		Li_2CO_3, H_3BO_3	4.1×10^{-6}b	$t_{1/2}$ = 112 min	*25*
F^{19}		Teflon	2.2×10^{-2}		
Al		Metal foil	8.4×10^{-3}		
Cl		NH_4Cl	2.2×10^{-3}		
Cu		Metal foil	8×10^{-5}		
Ag		Metal foil	1.6×10^{-6}		
Au		Metal foil	4.4×10^{-6}		
Cu	370	Metal foil	3×10^{-5}		*26*

649 ± 9 kev for the β^+ decay of F^{18}. These investigators also found the ground state of F^{18} to be $^3S_1$, and the disintegration energy Q_{β^+} = 1.67 Mev. The annihilation gamma was found by Knight, Novey, Cannon and Turkevich (*15*) to be 0.56 Mev.

Radiochemical Properties

Elemental fluorine occurs in nature as F^{19}. Three radioisotopes have been prepared. Their nuclear properties are listed in Table 2. The only convenient isotope for tracer studies is the longest-lived nuclide, F^{18}. Its annihilation gamma rays [0.56 Mev (*15*)] are readily counted, so that radio-assay is fairly simple. Fluorine-18 has been counted in the form of a solid, PbClF (*15*), a liquid, HF (*33*), and various gases such as HF, ClF_3, BrF_5, IF_7, and F_2 (*34*).

Knight *et al.* (*15*) precipitated radiofluoride ion from aqueous solution by the addition of HCl and $Pb(CH_3COO)_2$. The PbClF precipitate was washed, centrifuged and counted by conventional methods.

There has been little attention given to the problem of precise radio-assay of F^{18} in the liquid phase. Rogers and Katz (*33*) used an aluminum vessel to contain samples of liquid hydrogen fluoride under its vapor pressure at room temperature. A thin-walled mica end-window counter was placed adjacent to the bottom (0.010 in. thick) of the chamber. Difficulty in reproducibly filling the sample holder was probably the major factor responsible for the poor precision.

Most of the precise counting of F^{18} has been done in the gas phase. Dodgen and Libby (*35*) used a counter with a 0.010 in. aluminum wall contained in a brass jacket, sealed with Teflon gaskets. Into this counter gaseous hydrogen fluoride or fluorine was admitted. The gas volume was 67 ml. The pressure in the counting chamber was measured with a bronze tube Dura gage or a brass diaphragm "click gage." Counting rates of the order of 1,000–5,000 cpm for gas samples at 0.5 atmospheres were customarily obtained. All activities were corrected for decay, back to a convenient initial time, using a half-life of 112 min. Individual values of specific activity were quoted with an uncertainty of about ±2%.

Rogers and Katz (*33*) assayed F^{18} in the form of gaseous HF, ClF_3 and BrF_5. The gases were contained in a copper vessel (600 ml) in which a Victoreen 1B85 aluminum Thyrode was situated (Fig. 1). A vacuum-tight seal was made between the Thyrode and the copper vessel with a Fluorothene gasket. Lacquer was removed from the outside of the counter tube, and a thin layer of Fluorothene grease was used to protect the soldered joint at the end of the tube. The life of a given counter tube was approximately one month under continuous operation with fluoride vapors.

Weighed samples of radioactive HF, ClF_3 or BrF_5 were introduced into the chamber for determination of specific activity. For gas samples at pressures in the region of 1 atmosphere, counts of the order of 5,000–10,000 cpm were obtained. A small loss in handling was always observed.

One of the important sources of error in the radio-assay was the rather high

background in the counter due to the exchange of F^{18} between radioactive HF and the surfaces of AlF_3 and CuF_2, necessarily exposed to the gas during the counting periods. Because of this and other factors, the reliability of the determinations of specific activity was believed limited to about ±4%.

Bernstein and Katz (*34*) used essentially the same technique for the assay of HF, F_2, ClF_3, BrF_5 and IF_7, except that activities were computed on the basis of pressures rather than weights of the gaseous fluoride. Because of polymerization in the case of hydrogen fluoride, it is necessary to take into account the strong dependence of the molecular weight upon pressure and temperature (*36*). For ClF_3 a small correction for dimerization (*37*) is necessary. Figure 2 shows a typical decay plot for F^{18} in the form of ClF_3. The original pressure of ClF_3 in the counting vessel was 500 mm Hg.

After following the decay for approximately one half-life, the ClF_3 was removed from the chamber, which was then evacuated for background count. Then a portion of the ClF_3 was returned to the counting vessel (to a pressure of 250 mm) and counting resumed. No significant change in specific activity occurred. Figure 3 shows typical decay plots for F^{18} in HF, ClF_3 and BrF_5. Similar behavior was observed with IF_7 and F_2. Half-lives of 112 ± 1 min were consistently obtained for F^{18} regardless of the carrier species.

Adams, Bernstein and Katz (*38*) employed a nickel counting chamber; this appreciably reduced the background ascribed to the heterogeneous exchange with the surface. Evacuation of the counting vessel removed more than 99% of the activity. By the use of Fluorothene wax over the solder joint on the 1B85 Thyrode, the tube life was extended significantly. Gas pressures were measured to ±0.2 mm with a nickel diaphragm Booth-Cromer (*39*) null balance gage with an external pneumatic balancing system. Counting rates were 3,000–10,000 cpm, with 1-min counts taken at 5-min intervals. Specific activities were obtained from consideration of a large number of points along a decay curve, thus improving the statistical accuracy. By careful attention to detail, it was possible to attain a precision of ±1% in specific activity.

No attempt has been made to improve the geometry of the F^{18} counting chamber. For work involving a lower level of activity, this would seem to be quite necessary, since the relatively short half-life limits the period of counting and thus the statistical accuracy of the assay. Reduction of the annular gas space to a minimum volume would probably be the simplest method of increasing the efficiency. Further developments would be very valuable in extending the scope of investigations with F^{18}.

TABLE 2—Nuclear Properties of Fluorine Isotopes

Isotope	*Energy of radiation (Mev)*	*Ref.*	*Half-life*	*Ref.*
F^{17}	1.72 β^+	*30*	70 s	*30*
F^{18}	0.649 β^+	*23*	112 m	*38*
F^{19}	Stable			
F^{20}	5.41 β^-, 1.631 γ	*31*	10.7 s	*32*

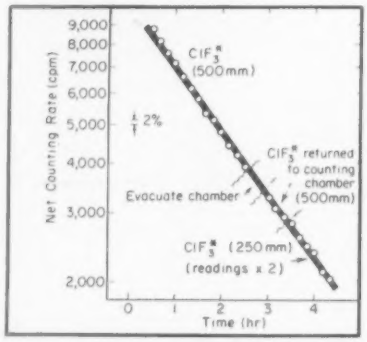

FIG. 2. Suitability of chlorine trifluoride for counting of fluorine-18

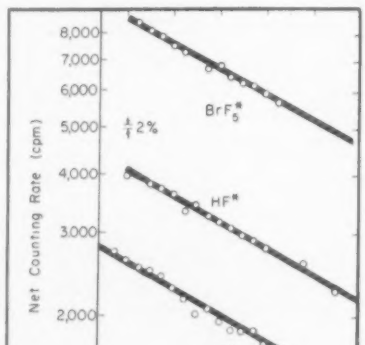

FIG. 3. Counting of fluorine-18 in various gases

Preparation and Handling of F^{18}

Of the large number of nuclear reactions that lead to the production of F^{18} (Table 1) only three general methods have proved convenient for routine application. The nuclear reactions are listed in Table 3 together with other relevant information.

For most applications any of the three methods of preparation are acceptable. Activities of 5,000–20,000 cpm are easily obtained. The choice would be dictated by the availability of equipment. For production of carrier-free F^{18}, the third method is required. Here one irradiates a sample of about 1 gm of $LiNO_3$ or $LiAlO_2$ in the thermal neutron beam of a nuclear pile for 1–8 hr. After a short cooling period the sample may be put into solution, giving carrier-free radiofluoride ion at a fairly high level of activity.

Radiochemical purity of the F^{18} is extremely high for the first and second methods, which are primary reactions of the fluorine with the incident photons and fast neutrons, respectively. However, the third method is essentially a secondary reaction, and radioactive contamination is serious unless stringent precautions are observed regarding purity of the materials irradiated. Knight, *et al.* (*15*) calculated that the yield of F^{18} from $LiNO_3$ is approximately 2×10^{-5} F^{18} nuclei per triton produced from the $LiNO_3$. Thus, any trace impurities, even of small thermal-neutron cross-section, would be capable of introducing very large spurious activities relative to the F^{18} produced by the triton reaction.

Adams, Bernstein and Katz (*38*) found that chlorine and bromine were the most serious contaminants from the viewpoint of radiochemical purity.

In one experiment, an intimate, equimolar mixture of LiF (Baker and Co.), containing 0.002% Cl, and Al_2O_3 was irradiated in a nuclear reactor for two hours. Figure 4 shows data on the decay of the contaminated ClF_3. The ratio of the Cl^{38} to F^{18} activities, extrapolated back to the time of removal of the sample from the pile, was about 0.2. To minimize this radiocontamination, materials containing <0.002% Cl were employed, and after an 8-hr bombardment the samples were given a cooling period of 4 hr to allow for decay of the Cl^{38}. Figure 5 shows a

decay curve extended over four half-lives of F^{18}, indicating satisfactory radiochemical purity, with a half-life of 112 ± 1 min.

Radiobromine isotopes (80 and 82) formed from trace impurities of Br presented a lesser problem. However, after 8–10 hr the contribution from Br^{82} (34 hr) was a perceptible fraction of the F^{18} activity. Fortunately, most of the experiments with radiofluorine are necessarily completed in less than four half-lives.

There was little success in reducing the radioactive halogen impurities by chemical or physical methods either before or after irradiation. Despite these complications, the nuclear reactor is a very convenient source of F^{18}, and virtually essential for studies involving carrier-free F^{18}.

Incorporation of F^{18} into various molecules. In the case of the first and second methods, the irradiated samples containing F^{18} in a carrier of fluoride may be put into aqueous solution, giving a tracer of radiofluoride ion. Preparation of labelled salts may be accomplished by conventional wet methods.

Much of the attention in the literature, however, has been centered on the preparation and reaction of labelled gaseous and liquid fluorides, such as hydrogen fluoride, fluorine and the halogen fluorides.

The conversion of labelled KHF_2* to HF* is readily accomplished by heating to 400° C in a nickel or copper vessel, previously pretreated with HF or F_2. The HF* may be transferred by trap-to-trap distillation. It is readily absorbed by anhydrous NaF pellets at 100° C; upon re-heating to 400° the $NaHF_2$* is decomposed, and HF* of half the original specific activity is recovered. Isotopic exchange between gaseous or liquid HF and the solid fluorides and bifluorides is apparently rapid and complete (*33*).

Two methods have been used for the preparation of labelled halogen fluorides. The halogen fluoride may be exchanged with HF* in the liquid or gaseous state. The gas-phase exchange reaction has been found to reach equilibrium in less than 3 min; equilibrium in the liquid phase is attained in less than 10 min (*33*). After mixing, the HF* is removed by fractionation, where feasible, followed by passage through a column of NaF pellets. The labelled halogen fluoride may be further purified by trap-to-trap distillation, using suitable temperature baths, with retention of the center cut.

Radioactive halogen fluorides may also be prepared by exchange with the LiF^*-Al_2O_3 sample. The halogen fluoride, preferably liquified under its own vapor pressure, is allowed to remain in contact with the LiF^*-Al_2O_3, previously outgassed, for 10–20 min (*34*). Exchange is accelerated by warming to 50°–100° C. Due to the extreme reactivity of the halogen fluorides, some noncondensible gas (presumably O_2) is usually formed during the time of exchange. This must be removed, and then the labelled halogen fluoride fractionated by trap-to-trap distillation. In the case of BrF_5, some difficulty was encountered because of a brown impurity, presumed to be BrF, which arose during exposure to the LiF-Al_2O_3 mixture. To remove this completely, the sample is treated with elemental fluorine and distilled.

To obtain radioactive elemental fluorine, the gas-phase exchange reaction between labelled HF or one of the halogen fluorides is utilized. These exchanges are rapid and complete (*34*) at temperatures in the neighborhood of 250° C, at pressures of 1–2 atm. The reactions are preferably carried out in pre-fluorinated nickel vessels.

For experiments involving weighed samples of HF or halogen fluorides, a small Fluorothene ampoule was constructed using a special small Crane bellows valve. The ampoule, weighing only 20 gm, was capable of withstanding pressures up to 2 atm.

Applications of F^{18}

It is somewhat surprising, considering the importance and wide-spread ramifications of fluorine chemistry, that so few investigators have availed themselves of the existence of this nuclide, known for 15 years. In this section, studies in which F^{18} has been employed will be reviewed, and an attempt to indicate other potential applications will be made.

Exchange studies. Libby and Dodgen (*35*) appear to have been the first to have used F^{18} in chemical exchange studies. These investigators were concerned with the general problem of halogen exchange in the gas phase between halogen acids and halogens (HX-X_2), and to this end, among other exchanges, studied fluorine exchange in the HF^*-F_2 system. These results were of a preliminary nature, and indicated that the exchange observed at moderately elevated temperatures was probably heterogeneous in nature, occurring on the walls of the brass container used. Their conclusion was fortified by the observation that exchange occurred between HF* and the fluoride film on the walls of the reaction vessel. The possible existence of a homogeneous gas-phase exchange in addition to the heterogeneous reaction could not be excluded on the basis of their results.

An attempt was made to induce the exchange at room temperature by photochemical generation of F atoms. A cell with fluorothene windows was filled with approximately equal amounts of HF and F_2. After illumination for 30 min with the full radiation (3,650 Å) of a mercury arc, negligible exchange had occurred. It was estimated that 4×10^{12} F atoms/cm³/sec were generated. The cell contained 10^{19} F_2 molecules per cm³. Further work on this interesting system is clearly indicated.

TABLE 3—Preparation of F^{18}

Nuclear Reaction	*Irradiation*	*Target*
$F^{19}(n,\gamma)F^{18}$	Betatron 48–84 Mev X-rays	KHF_2(*33*) or any fluorine containing substance
$F^{19}(n,2n)F^{18}$	Cyclotron Fast neutrons from $Li^6(d,n)Be$	HF, KHF_2, etc.
$O^{16}(t,n)F^{18}$	Nuclear reactor 2.65-Mev tritons from $Li^6(n,\alpha)H^3$	$LiNO_3$,(*15*) $LiAlO_2$,(*34*) LiF + Al_2O_3.(*38*) Any comb. of Li and O satisfactory

These exchange studies have recently been extended to fluorine exchanges between hydrogen fluoride and the halogen fluorides ClF_3, BrF_3, BrF_5, IF_5, and IF_7. Emeléus and co-workers (*2*) have shown that BrF_3 and IF_5 undergo extensive self-ionization. Chlorine trifluoride and bromine pentafluoride show little tendency to self-ionization, but participate in ionic equilibria

$$ClF_3 + HF \rightleftarrows ClF_2^+ + HF_2^- \quad (1)$$
$$BrF_5 + HF \rightleftarrows BrF_4^+ + HF_2^- \quad (2)$$

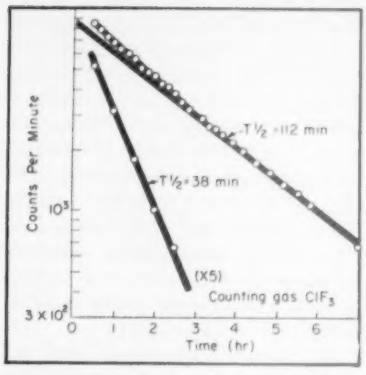

FIG. 4. Typical decay curve of F^{18} showing contaminating 38-min Cl^{38} activity

Similar ionic equilibria have been postulated for other halogen fluorides

$$BrF_3 + HF \rightleftarrows BrF_2^+ + HF_2^- \quad (3)$$

$$IF_5 + HF \rightleftarrows IF_4^+ + HF_2^- \quad (4)$$

If these equilibria do in fact exist in mixtures of hydrogen fluoride and the halogen fluorides, then exchange of radioactive fluorine should occur with great readiness via the common ions. It has actually been observed (*33*) that HF* exchanges fluorine completely in 10 min or less with BrF_3, ClF_3, BrF_5, IF_5 or SbF_5. All the fluorine atoms in the halogen fluoride were exchangeable.

When similar exchanges were examined in the gas phase (*33*), it was found that HF* exchanges fluorine rapidly with ClF_3, BrF_3, BrF_5, and IF_7 at room temperature. It is not known whether these exchanges are homogeneous or heterogeneous reactions. If they are homogeneous, then a number of interesting problems are posed regarding the nature of the activated complexes involved in the exchange. Similarly, ClF_3* and BF_3 were found to exchange very rapidly in the gas phase. With fluorine compounds in which the bonds are predominately of covalent character such as SF_6 or CCl_2F_2, no exchange was observed with HF*.

Bernstein and Katz (*34*) studied the gas-phase exchange of F^{18} between elemental fluorine and the halogen fluorides ClF_3, BrF_5, and IF_7 in vessels of copper, aluminum and nickel.

Equilibrium was attained only at elevated temperatures (300° C). Under these conditions, it was shown quantitatively that all the fluorine atoms of the halogen fluoride are exchangeable with elemental fluorine. Conveniently measurable exchange rates were obtained between 120–250° C.

Although the data showed that the nature of the surface affected the rate, it was considered that the exchanges were occurring primarily by a homogeneous mechanism. This mechanism was based on the reversible dissociation of ClF_3 and IF_7. However, in the case of BrF_5, where no measurable dissociation occurs up to 400°, this mechanism is unacceptable. The possibility of an associative mechanism for this exchange was suggested.

A general formula for the exchange rate was derived, using McKay's method (*40*)

$$R = \frac{2nab}{(na + 2b)t} \ln\left(\frac{1}{1-f}\right)$$

where R is the rate of the exchange reaction, f the fraction exchanged in time t, a the concentration of the halogen fluoride XF_n, b the concentration of fluorine, and n the number of exchangeable atoms of F in the halogen fluoride.

A quantitative study of the kinetics of these exchange reactions was later carried out by Adams, Bernstein and Katz (*38*). It was found that the rate was a linear function of the concentration of F_2, and a complex function of the concentration of the halogen fluoride. In the case of IF_7, the results could be interpreted, to a first approximation, on the basis of a homogeneous, dissociative mechanism. For the BrF_5-F_2 exchange, where this mechanism was excluded, it was necessary to postulate a heterogeneous mode of exchange, with the NiF_2-coated surface of the exchange vessel acting as the catalyst. If one assumed that F_2 was weakly adsorbed and that BrF_5 was moderately adsorbed on the surface, the observed dependence of the rate upon the concentrations of F_2 and BrF_5 was obtained. The mechanism implied competition for surface sites by F_2 and BrF_5, similar to that postulated for the NH_3-D_2 exchange on Pt (*41*). The kinetics of the ClF_3-F_2 reaction was found to be intermediate between the cases of IF_7 and BrF_5. A combined homogeneous and heterogeneous mechanism was invoked to explain the data. Unfortunately, experiments with added surface failed to demonstrate the proper acceleration of the exchange rate, so that the details of the mechanism are not yet understood.

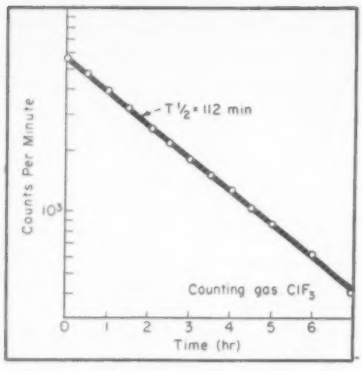

FIG. 5. Half-life of F^{18} produced by neutron bombardment of LiF and Al_2O_3

Further exchange studies would appear to be needed. Such systems as F_2-ClF, F_2-BrF_3, F_2-IF_5, F_2-SF_6, F_2-BF_3, F_2-MoF_6, and others offer interesting possibilities. Exchange reactions among the interhalogen compounds in the gas phase would aid in the clarification of the mechanism of the HF-XF_n and F_2-XF_n exchanges. In addition, the kinetics of the exchange of HF and F_2 remain to be investigated thoroughly.

Other interesting fluorine exchange studies suggest themselves in the field of aqueous solutions. The exchange between F^- and complex ions of fluorine such as FeF_6^{---} would seem to be worth investigation. The problem of separation would be the major one in such studies. Ion exchange techniques might be useful here.

The interaction of gaseous and solid fluorides has been subjected to cursory examination by several authors (*33*, *34*, *35*). Table 4 summarizes some of the conclusions based on these studies. Conditions were not always comparable, unfortunately, so that the results have only qualitative significance.

Bernstein and Katz (*34*) reported data on the exchange of ClF_3 vapor with CaF_2 of known surface area. Approximately two layers of fluoride ions on the surface of the CaF_2 crystal were found to undergo exchange with the ClF_3. Further work would be required to ascertain whether or not it would be possible to measure the surface area of solid fluorides by the exchange method.

Biological investigations with F^{18}. The role of fluorine in human dentition is at present under very active investigation by many workers; the first ap-

plication of F^{18} was in connection with this problem. In 1940, Volker, Hodge, Wilson and Van Voorhis (*42*) studied the adsorption of fluoride ion by enamel, dentin, bone and hydroxyapatite. The F^{18} used in this work was prepared by irradiation of water using a proton beam from a cyclotron: $O^{18}(p,n)F^{18}$. As might be anticipated, only a relatively few counts of F^{18} could be obtained. The adsorption of F^- was readily determined by noting the amount of activity removed when the solution was equilibrated with the various solids. The adsorption was found to follow the Freundlich adsorption isotherm.

In 1941, Volker, Sognnaes, and Bibby (*43*) extended this work to a study of the distribution of fluoride in the bones and teeth of experimental animals. Radioactive fluorine, prepared as above, was given by interperitoneal injection to rats and by intravenous injection to cats. The failure to find significant amounts of radioactive fluoride in dental tissues well removed from the tooth pulp led these investigators to conclude that it was quite improbable that fluorine is deposited in dental enamel through the blood stream.

Very recently, Myers, Hamilton, and Becks (*44*) have studied the topical application of fluoride to teeth with F^{18}. Tracer was prepared by bombardment of triply distilled water with alpha particles accelerated in the Berkeley cyclotron; the nuclear reaction is $O^{16}(\alpha,np)F^{18}$. The irradiated water is rendered slightly alkaline to prevent loss of fluoride, and sodium fluoride added. This solution is then evaporated to yield a 2% NaF solution.

After topical application, about 2% of the fluoride is transferred to the tooth; the major portion of this is removed on rinsing. An autoradiograph showed that F^- concentrates in surface defects and discolored enamel of the crown. These investigators concluded that the protective action of topical fluoride is related to uptake by imperfections.

The possibility of studying the binding of F^- by isolated enzyme systems, such as in the carbohydrate metabolic system that is known to be strongly inhibited by fluoride ion, also suggests itself.

Miscellaneous applications. At the present time, no entirely satisfactory analytical method for the determination of fluoride is known. F^{18} may be of use in this connection in several ways.

First, the method of isotope dilution could be applied to this determination. Second, F^{18} could be used as a tracer to aid in the evaluation of any proposed new analytical techniques, such as precipitation, complex formation, and ion exchange.

To use F^{18} as a tracer in the development of better analytical methods, one needs only to study the relative efficiency of the separation procedure as the experimental conditions are changed, working from aliquots of a stock solution of labelled fluoride ion. This technique is ideally suited to studies of ion exchange methods of separation, using conventional tracer techniques for evaluation of the resin capacity, the rate of uptake, and the rate of elution of fluoride ion. No experimental work of this kind has been reported up to the present time.

Solubilities of a large number of fluorides in anhydrous hydrogen fluoride are known (*45*). Studies of this kind might well be extended to substances of considerably lower solubility and to lower temperatures by the use of F^{18}-labelled compounds, readily tagged by either of the first two methods previously described (Table 3).

Rates of dissolution might be measurable by taking advantage of the convenient gas-phase radioassay of HF* provided the rate of equilibration of F^{18} between liquid and gaseous HF was greater than the rate of dissolution.

Another application of F^{18} might be in connection with fundamental studies involving the chemistry of the Freons, used extensively as refrigerants. Labelled Freons may be readily prepared by γ-ray or fast-neutron bombardment of the appropriate compounds.

TABLE 4—F^{18} Exchange between Gaseous and Solid Fluorides at Room Temperature (*33, 34*)

	NaF	*LiF*	*$NaHF_2$*	*CaF_2*	*CuF_2*	*NiF_2*	*AlF_3*
HF	+++	+++	+++	+	+	+	+
F_2	–	–	–	–	–	–	–
ClF_3	++	+++	++	+	+	+	+
BrF_3	++	+++	++	+	+	+	+
IF_7	++	+++	++	+	+	+	+

Symbols: +++ rapid and complete exchange with bulk solid.
++ slow exchange with bulk solid.
+ exchange only surface layers of solid.
– negligibly slow or no exchange.

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Radiophotoluminescence Dosimetry System of the U. S. Navy

This dosimeter, utilizing Ag-activated phosphate glass, has a range of from 10 r to at least 600 r. Response is linear and independent of dose rate up to at least 170 r/min. Energy dependence is diminished by lead filters

By JAMES H. SCHULMAN,* WILLIAM SHURCLIFF,† ROBERT J. GINTHER,* and FRANK H. ATTIX‡

THE ADVENT OF NUCLEAR WEAPONS has created a need for small, inexpensive, and rugged high-range gamma-ray dosimeters for general distribution to military and civilian personnel. This need is satisfied to a considerable degree by the DT-60/PD personnel dosimeter developed under the auspices of the U. S. Navy Bureau of Ships, utilizing the radiophotoluminescence system of dosimetry proposed at NRL in 1947 (*1, 2, 3*).

The DT-60/PD, about the size of a pocket watch and weighing slightly less than an ounce, can easily detect as little as a 10-r dose, and registers doses well beyond the lethal range. It survives Navy shock, immersion, and temperature tests, and has an indefinitely long shelf life. Its present cost is less than one dollar. The DT-60/PD is not a self-indicating device, but is easily read in a few seconds with the aid of a fluorimeter (the CP-95/PD Radiac Computer Indicator).

Radiophotoluminescence (*4, 5*) may be defined as the creation of new stable photoluminescent centers in a material by the action of ionizing radiations. After exposure to such radiations, a radiophotoluminescent material will fluoresce under light of a properly selected (usually near-ultraviolet) spectral region, while undosed material will not fluoresce under this same light.

The intensity of the fluorescence, under a fixed intensity of excitation, is proportional to the dosage (*3, 5*). The radiation-induced photoluminescence is, ideally, a permanent property of the material; the luminescence continues undiminished in intensity, without disturbing its dosage indication, as long as the material is illuminated by the exciting light. In this important respect, radiophotoluminescence differs most significantly from radiophotostimulation, and radiothermoluminescence, which can serve as bases for other dosimetry systems (*5, 6, 7*). These phenomena involve the production of a temporary luminescence through optical or thermal release, respectively, of the energy stored in irradiated phosphors.

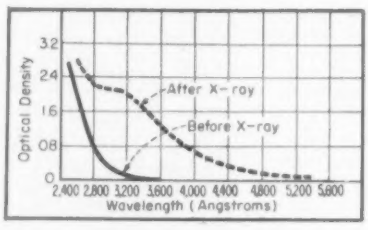

FIG. 1. Absorption spectra of Ag-activated phosphate glass

Ag-Activated Phosphate Glass

From the various materials that were found to be radiophotoluminescent (*3, 4, 5, 8*), a silver-activated phosphate glass* was selected, based on considerations of low cost, ease of large-scale manufacture, and insolubility in water and other solvents.

The glass is transparent throughout

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† Polaroid Corp., Cambridge, Mass.
‡ National Bureau of Standards, U. S. Dept. of Commerce, Washington, D. C.

* This glass, developed by W. A. Weyl (*9*), has the following composition by weight: 50% $Al(PO_3)_3$, 25% $Ba(PO_3)_2$, and 25% KPO_3. Up to 16% by weight of $AgPO_3$ can be incorporated in this base glass. A silver content corresponding to about 8 weight per cent of $AgPO_3$ was most satisfactory from the standpoint of sensitivity and other properties (*1, 5*).

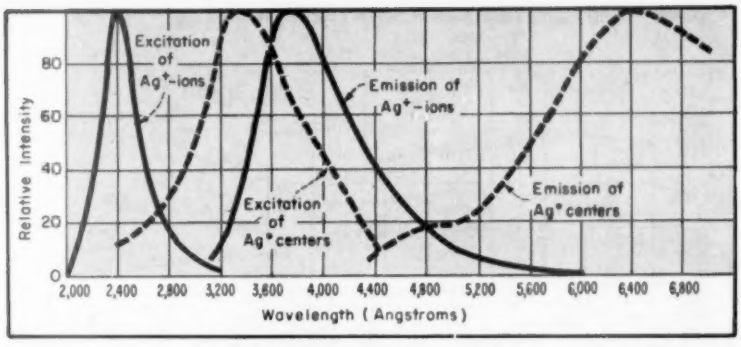

FIG. 2. Spectra of undosed (Ag^+ centers) and irradiated (Ag^0) phosphate glass

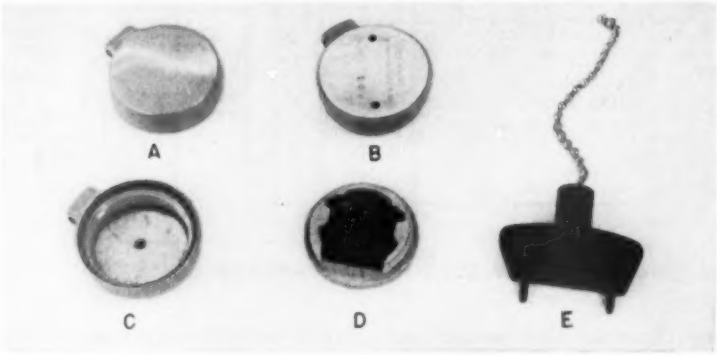

FIG. 3. DT-60/PD dosimeter: (a) assembled dosimeter, top side, and (b) underside; (c) cover portion, showing lead shield; (d) base portion, showing glass block; (e) wrench for assembly and disassembly of dosimeter

the visible spectrum and in the near ultraviolet down to its absorption edge at about 3,000 Å (Fig. 1). Under excitation by short-wave ultraviolet (2,537 Å), the undosed glass emits a broad fluorescence band peaking in the violet and extending over a wide portion of the visible spectrum (Fig. 2). This luminescence is due to the Ag^+ ion. Under near-ultraviolet light, about 3,500 Å, the luminescence of the undosed glass is very feeble, although not zero. This weak luminescence may likewise be due to Ag^+ ion, or it may be due to traces of luminescent impurities in the glass.

After dosing with X-rays or γ-rays, a new, poorly resolved absorption band appears on the long-wavelength side of the original absorption edge of the glass (Fig. 1). The new band peaks at 3,200 Å, overlaps the original glass absorption band and the Ag^+ ion excitation band at shorter wavelengths, and extends into the visible spectrum on the long-wavelength side.* Excitation by light lying in this new absorption band produces an orange luminescence (Fig. 2).

Electrons liberated by the X-rays are trapped by the Ag^+ ions of the glass, reducing them to "atomic" silver centers, Ag^0, which are responsible for the new absorption band (*8*). Light absorbed by the Ag^0 centers does not destroy them, but merely raises them to an excited state, from which they return to the ground state with the emission of the orange fluorescence.

If the dosed glass is observed under ultraviolet light common to the excitation bands of both Ag^+ and Ag^0, say 3,000 Å, the resulting fluorescence will be composed of two emission bands. To isolate the Ag^0 fluorescence as much as possible—for it is the intensity of this fluorescence that is proportional to the dosage—longer-wavelength ultraviolet, near 3,600 Å, is used for excitation, and the glass is observed through an orange filter. This arrangement minimizes the "predose"† signal from the Ag^+ ions, and emphasizes the orange fluorescence of the Ag^0 atoms.

Dosimeter and Fluorimeter‡

The DT-60/PD dosimeter, shown in Fig. 3, consists of a $\frac{3}{4} \times \frac{3}{4} \times \frac{3}{16}$ in. block of silver-activated phosphate glass encased in a circular plastic locket 1½ in. in diameter and ½ in. thick. Ethyl cellulose was found to be very satisfactory as the plastic.

The glass is rough ground, and painted black on all but one $\frac{3}{4} \times \frac{3}{4}$ in. face (the "ultraviolet entrance face") and one $\frac{3}{4} \times \frac{3}{16}$ in. face (the "fluorescence exit face"). The simple shape and rough finish of the glass were chosen in the interest of economy.

The locket is constructed in two screw-threaded parts, a "base" and a "cover." To correct for the energy dependence of the glass, a circular piece of 0.046-in.-thick lead, containing a 0.107-in.-diameter central hole, is cemented to the inside of the cover. A similar $\frac{3}{4} \times \frac{3}{4}$ in. square piece of lead is cemented to the inside of the base. The glass is, in turn, cemented to this square lead shield, with the large painted face of the glass bonded to the lead. A part of the male threading of the locket base is cut out, exposing the fluorescence exit face of the glass. A gasket is provided to make the closure between base and cover watertight. Two holes, located in the back on the base, serve as grips for a two-prong wrench used in opening and closing the locket. In reading the dosage registered, the cover is unscrewed and only the base portion is inserted into the fluorimeter.

Fluorimeter. Two models of the fluorimeter or "reader" have been developed, one, the CP-95(XN-3)/PD, for precise laboratory and control work (Fig. 4), and the other, the CP-95/PD, for field use (Fig. 5).

The operation of both instruments is very similar. Near-ultraviolet light, in the region of 3,650 Å, impinges perpendicularly on the ultraviolet entrance face of the glass. This light is provided by an RP-12 "blacklight" lamp fitted with a Corning 5860 filter to remove the visible light also emitted by the lamp. Fluorescent light emerges from the fluorescence exit face of the glass block and, after passage through

* Because of the long-wavelength extension of this band, high doses of radiation color the glass yellow.

† The fluorescence of an undosed dosimeter (due to Ag^+ ions and impurities) is hereafter called its "predose" fluorescence. The total observed fluorescence of an irradiated dosimeter (due to Ag^+ ions, impurities, and Ag^0 atoms) is called "postdose" fluorescence.

‡ Design features of these devices and other features of the dosimetry system are covered by patents pending.

FIG. 4. CP-95(XN-3)/PD reader

FIG. 5. CP-95/PD field reader

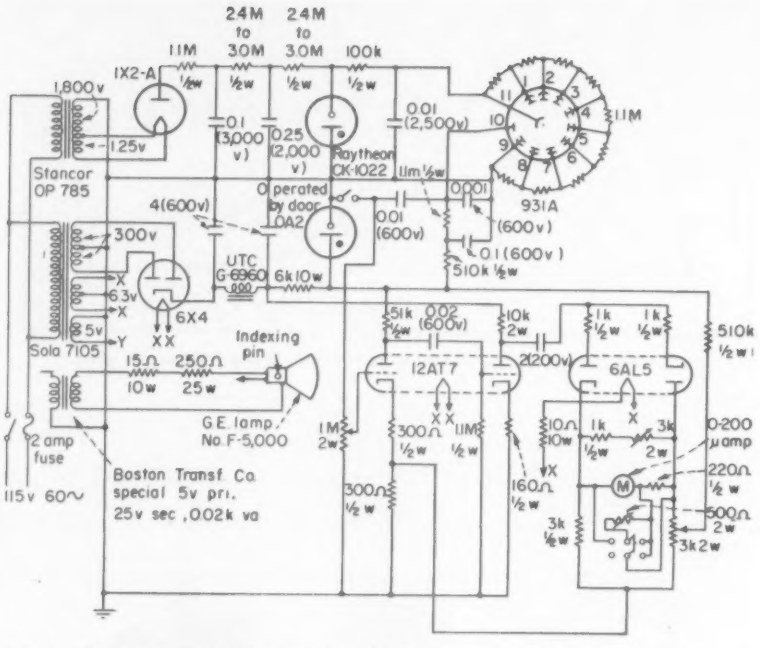

FIG. 6. Circuit diagram of the CP-95(XN-3)/PD laboratory reader

a Corning 3482 (orange) filter, impinges on the cathode of a 931A photomultiplier. Indication is provided by a meter calibrated to read directly in roentgens. The photomultiplier voltage may be varied to compensate for variations in lamp output.

The means for positioning the DT-60/PD inside the reader consists of a horizontal sector with wells and locating pins to accept the dosimeter. By means of an external lever, the sector may be swung out of the body of the instrument to permit insertion of a dosimeter, the light-tight front door of the reader being automatically opened by this motion. After loading, the sector is swung back inside the instrument for reading, and the door automatically closes behind it. Accurate positioning of the sector inside the instrument is effected by means of detents.

The laboratory reader measures $9 \times 10 \times 15$ in., weighs 28 lb, and operates on 110 volts, a-c, consuming 35 watts. The field instrument measures $16\frac{1}{2} \times 8\frac{5}{8} \times 9\frac{1}{4}$ in., weighs 24 lb, and operates on 110 volts, a-c or d-c, dissipating 36–38 watts. Both instruments have a 0–200 and a 0–600 r scale. The circuit diagram of the CP-95(XN-3)/PD is given in Fig. 6 and that of the CP-95/PD in Fig. 7. A battery-operated reader, using a pulsed ultraviolet light source and weighing about 7 lb, is under development at NRDL.*

Laboratory Operations

The predose fluorescence of the DT-60/PD's is first determined with the photomultiplier voltage control set at some convenient arbitrary value. A representative sample number of dosimeters are then given a known dose of X-rays or γ-rays, and their postdose fluorescence is then determined at the same control setting previously used. The sensitivity of the system (in scale divisions per roentgen) at this setting is

$$\frac{\text{(postdose reading)} - \text{(predose reading)}}{\text{(known dose delivered)}}$$

With this calibration, the dosage received by any dosimeter may be determined from its postdose and predose readings. Doses as little as 2 r can be detected by this procedure.

Field Operation

For field operations, where the ultimate in sensitivity and accuracy are not needed, a system has been set up whereby the dosage registered by an average production DT-60/PD may be determined from a single reading, with no calibration or computation by the operator of the reader.

This system takes advantage of the close control of predose values achieved in mass-production of the DT-60/PD. By appropriate methods of glass manufacture and by careful control of trace impurities, the predose fluorescence and the sensitivity of the glass can be varied within wide limits. In the interest of low cost and wide-spread availability of the glass-making raw materials, the predose-vs-sensitivity relationship chosen for production was such that a dose of 37 roentgens doubles the original fluorescence of the glass, i.e., the predose fluorescence is equivalent to a dose of 37 r. A large master batch of DT-60/PD's, made from glass carefully manufactured to these specifications, has been set aside as a primary standard for control of predose fluorescence and sensitivity.

With this close control, the rms variation in predose fluorescence is equivalent to 3–5 r, which is acceptable for field use. The field reader is adjusted to subtract the average predose reading automatically. This is accomplished by displacing the roentgen scale so that its zero is not coincident with the zero of the meter. In laying out the 0–200 roentgen scale, for example, the full meter deflection is taken as 237 units. The zero of the roentgen

* Designed by Mr. George Work of NRDL.

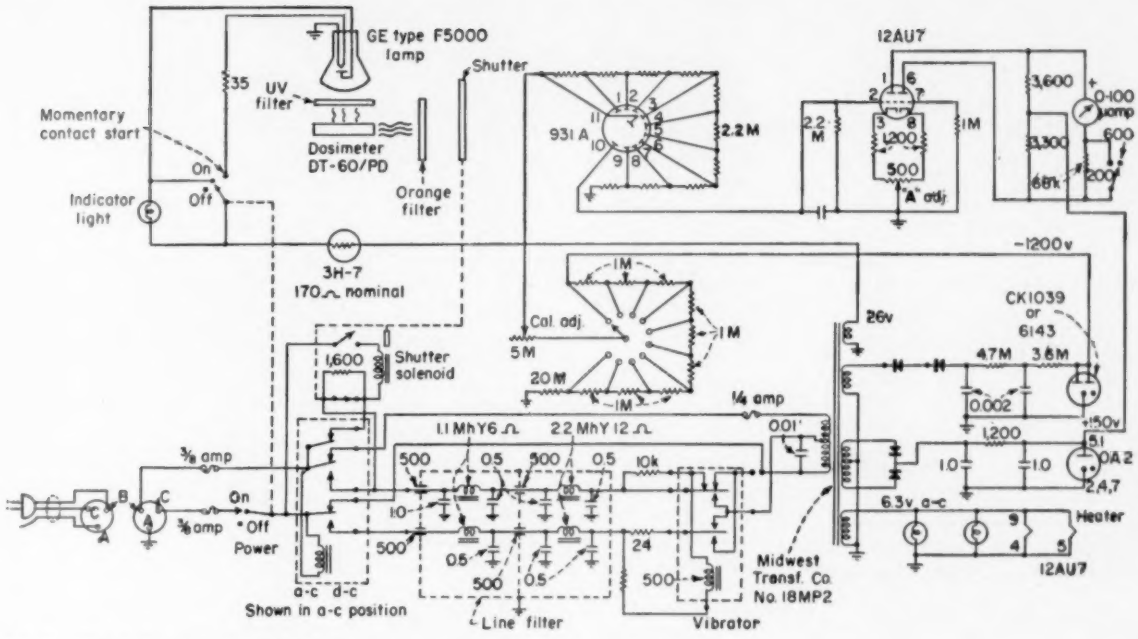

FIG. 7. Circuit diagram of the field reader

scale is marked at a point 37/237 of full scale, and full scale is marked 200 r. A single reading thus gives the dosage directly in roentgens.

Each field reader is provided with a permanently fluorescent glass standard built into the swinging sector of the instrument. This standard is made of a radiation-insensitive manganese-activated glass that fluoresces under near-ultraviolet light with an emission closely resembling that of the dosed silver-activated phosphate glass. The intensity of the fluorescence is adjusted so as to stimulate a dose of about 180 r, the exact value being stamped on each standard. Adjustment of the field reader is effected merely by setting the photomultiplier voltage so that the reading, with the standard in position, is the value specified on the standard.

Performance of the DT-60/PD

Growth and persistence of fluorescence-ability. Immediately after irradiation with a dose delivered in a short time interval, the measured fluorescence of the DT-60/PD dosimeter is 10–20% lower than the value it reaches after several hours.

The "build-up" is rapid at first, so that the reading after 2 hr is usually within 5% of the final value, and in about 24 hr the equilibrium value is reached. The build-up time is shorter for glass of higher silver content and longer for glass of lower silver content. With silver concentrations much greater than that of the composition selected, the predose fluorescence is undesirably high.

After 24 hr, the fluorescence-ability of the glass is constant, and has not been observed to change on storage for months at room temperature.

For the purpose of studying the other characteristics described here, the dosimeters were allowed to reach the full equilibrium value, being read one day after termination of the exposure.

Linearity of response. The fluorescence-ability of the DT-60/PD increases linearly with the applied dose, as indicated in Fig. 8.

Independence of dose rate. The fluorescence-ability produced in the dosimeter by a given total dose of radiation does not depend upon the rate at which the radiation is delivered for dosage rates at least as high as 170 r/min.

Additivity. Because of the stability of the fluorescence and linearity of the response of the DT-60/PD dosimeter, this dosimeter integrates a number of doses administered at intervals over a long period of time.

Energy dependence. The effect of gamma radiation upon the silver-activated phosphate glass depends greatly on the energy of the photons. Figure 9 shows that, for bare glass, 70-kev radiation is eighteen times more effective than Co^{60} gamma rays in producing fluorescence-ability. This energy dependence is diminished in the DT-60/PD by sandwiching the glass element between two lead filters within the plastic case. The resulting response of the dosimeter as a function of the energy of photons incident perpendicularly to the plane of the case is illustrated in Fig. 10. The increase in response in going from 120 kev down to 80 kev is brought about by the presence of the small hole in the center of each lead filter.* Without the hole, the lead absorption causes the trend in response from 180 to 120 kev to continue sharply downward, making the dosimeter very insensitive to lower-energy radiations.

Dependence on direction of incident radiation. As might be expected, considering the configuration of the lead filters in the dosimeter case, the DT-60/PD is more sensitive to radiation incident from some directions than from others. The magnitude of this

* This method of retaining response to softer radiations is due to Dr. Clifford C. Klick of the Naval Research Laboratory.

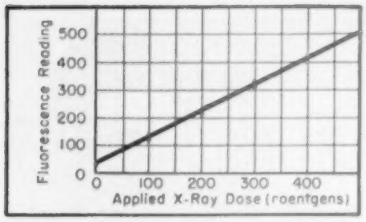

FIG. 8. DT-60/PD dosimeter response

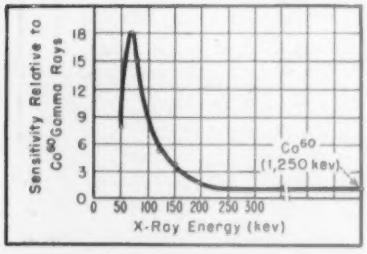

FIG. 9. Energy dependence of response of unshielded Ag-activated phosphate glass

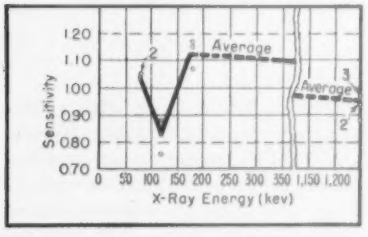

FIG. 10. Energy dependence of the DT-60/PD to radiation incident perpendicularly to plane of the case

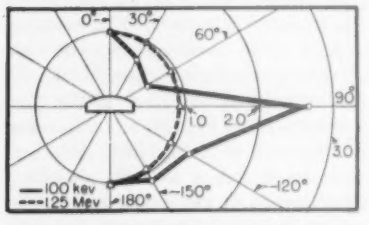

FIG. 11. Relative dependence of response of DT-60/PD dosimeters on direction of radiation (response at front of case, 0 deg, is taken as unity)

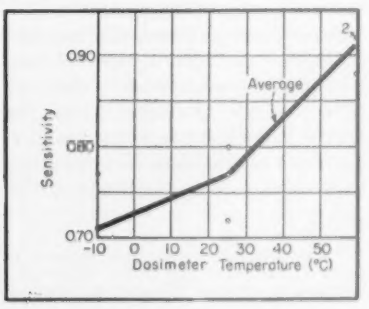

FIG. 12. Effect of temperature during irradiation on DT-60/PD response

directional dependence is greater at low energies than at high energies. Figure 11 shows this effect at two different energies, 1.25 Mev from Co^{60}, and 100 kev effective energy from an X-ray machine operating at 250 kv constant potential, with 1.7-mm Cu added filtration. Improvement of the directional dependence of the dosimeter should be possible by providing lead filters around the edges of the glass.

Temperature effects. The dosimeters are subject to three types of temperature effects. The DT-60/PD registers a higher apparent dose if it is kept at an elevated temperature while being exposed to gamma radiation (and is cooled to room temperature before being read). The effect of irradiation temperature is shown in Fig. 12. Some variation in the shape of this curve has been noted among different melts of glass, but the curve given is representative. Since the DT-60/PD, in its role of a personnel dosimeter, is worn near the body, its temperature during use is not expected to vary over the wide range shown in Fig. 12, and the irradiation temperature effect is therefore not expected to lead to appreciable errors.

The second type of temperature effect relates to the temperature at which the dosimeters are stored during the time period between exposure and fluorescence-reading (see Fig. 12 of ref. *5*). A longer time is required to attain the final value of the fluorescence reading when the dosimeter is stored at low temperatures than is required if the dosimeter is stored at room temperature, and the final fluorescence response is also somewhat lower than that achieved by room temperature storage. However, this effect, like the preceding one, is not important when the dosimeter is worn near the body as a personnel dosimeter.

The third type of temperature effect concerns the temperature of the DT-60/PD at the time it is being read. In common with all luminescent materials, the efficiency of luminescence of the dosed glass decreases with increasing temperature, the temperature coefficient of the glass being approximately $-0.7\%/°C$. By taking simple precautions to avoid temperature differences between the standard and the dosimeter during the reading, this small effect can be avoided entirely, if desired.

* * *

The development of this dosimeter system to production form has involved the direct participation of five government agencies and seven private companies. Several other organizations have served in an advisory capacity.

After demonstration of the feasibility of the system at the Naval Research Laboratory, using both visual and photoelectric fluorimetry, further evaluation tests were conducted by Raymond Alger of the Naval Radiological Defense Laboratory. Glass for these tests was prepared by the Naval Research Laboratory and by Norbert Kreidl of the Bausch and Lomb Optical Company.

Assuming direction of the development in 1950, the Bureau of Ships awarded a research contract (NObsr-42257) to the Polaroid Corporation, under which the present forms of the DT-60/PD dosimeter and the CP-95(XN-3)/PD reader were developed. Personnel from the Naval Research Laboratory, Bureau of Ships, and the National Bureau of Standards contributed significantly to the design of these units. Research contracts (NObsr-57010 and NObsr-67016) were awarded in 1951 to the Bausch and Lomb Optical Company for further studies of the sensitive glass. The permanently-fluorescent glass used as a standard in the field reader was developed by this company.

The first production contracts for the dosimeter were awarded in 1951 to the Polaroid Corporation and the Corning Glass Works. Later procurement contracts included the Penberthy Instrument Company as a participant in the production. The CP-95 (XN-3)/PD readers used for experimental, standardization, and control work were manufactured by the Polaroid Corporation. Development and production of the CP-95/PD field reader were both performed, under a production contract, by the Admiral Corporation, the development phase involving collaboration with the Naval Research Laboratory and the Polaroid Corporation.

Preproduction evaluations of the DT-60/PD models were carried out by the Material Laboratory of the N. Y. Naval Shipyard, the National Bureau of Standards, and the Naval Radiological Defense Laboratory. The preparation of standards used in coordinating the program are now being carried out at the last-named establishment.

The Pittsburgh Plate Glass Company and the Pilot Chemical Company have been connected with the program as suppliers of glass to the Polaroid Corporation.

In addition to the persons mentioned above and in the text, special acknowledgment is due to the following personnel, formerly of the Radiac Section of the Bureau of Ships: Joseph C. Cryden, the first project engineer in charge of the program; Charles R. Horner, who later assumed direction of the program; and to Adolph Lovoff, Chief of the Radiac Section during the early period of this work. Since May, 1952, Samuel C. Rainey has been the project engineer in charge of dosimeter work in the Radiac Section of the Bureau of Ships.

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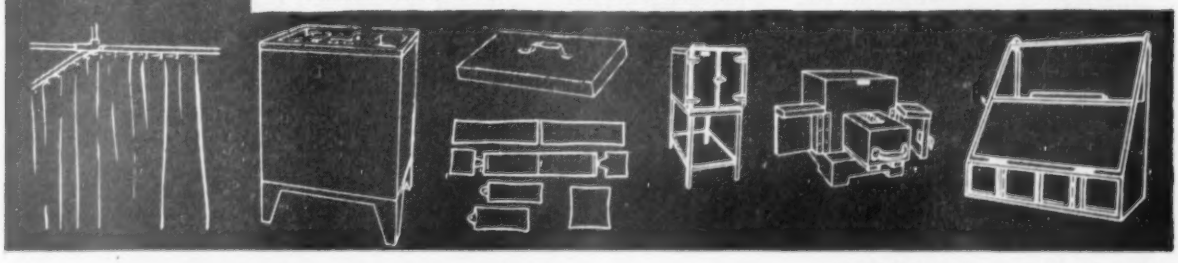

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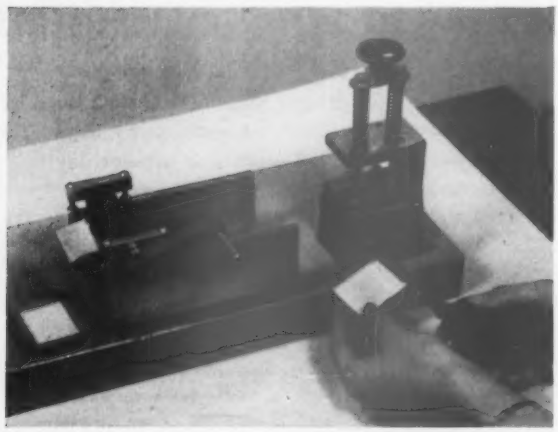


FIG. 1. Press used for forming sample cups (right). Pressure on bulb in operator's hand ejects sample cup. Flattened sample cup with Scotch tape cover is shown at left. Scotch tape edges are folded under before rolling (center)

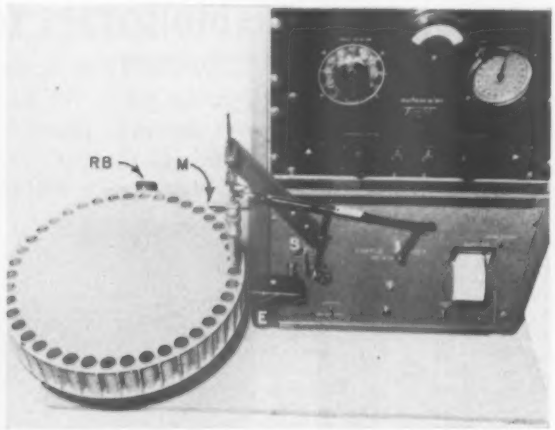


FIG. 2. Dipping-counter automatic sample changer is shown while changing samples. Brake release solenoid *RB* is energized and drive motor *M* is engaged. Geiger tube remains lifted until switch *E* stops turntable

Automatic Sample Changer and Recorder for Dipping Counters

By B. P. McKAY and D. B. ZILVERSMIT
Division of Physiology, University of Tennessee
Memphis, Tennessee

The advantages of automatic counting have been recognized by all investigators faced with a heavy counting load. Until recently the expense of commercial automatic sample changers has prevented many laboratories from using them. Even today there are only a few automatic sample changers commercially available, and all of them employ end-window Geiger tubes as sensing elements.

In our laboratory we have preferred the Victoreen 1B85 Thyrode in conjunction with flexible samples wrapped around the Geiger tube for automatic counting. A comparison of this method with the end-window procedure might be helpful in the evaluation of the sample changer to be described.

Comparison of Methods

In end-window counting the sample is commonly mounted on a planchet or dish. In the latter instance either liquid or dry samples can be used. Even though the use of liquid samples under an end-window tube is a time saving and convenient procedure, there are serious objections to this method when it is used indiscriminately. The loss of efficiency by the large self-absorption in the liquid sample is perhaps the least shortcoming of this procedure when used with hard beta or gamma emitters. Evaporation of the solvent before or during the counting can lead to erroneous results. Evaporation not only decreases the thickness of the sample, and thus its self-absorption, but it frequently leaves a rim of highly radioactive deposit at the top of the sample cup. Creeping of the radioactive solution to the top of the sample cup and even to the outside surface of this cup has been demonstrated in our laboratory when P^{32}-labeled phospholipides were counted in ether solutions by this method.

In biological work the liquid phase is frequently not homogeneous. Tissue digests, for example, may contain radioactive fat droplets that possess amounts of radioactivity different from the water phase. A nonuniform distribution of these fat droplets and a tendency to concentrate at the top of the sample during counting will certainly lead to erroneous results.

The opposite condition is met when whole blood is used for counting. Usually the red blood cells contain more or less radioactivity than the serum. Sedimentation of these cells during counting will thus affect the counting rate.

Even in purely inorganic solutions where particles are not visible to the naked eye or the microscope, migration of radioactivity is known to occur. We have observed that when radioactive gold foil is dissolved in aqua-regia, and subsequently diluted for counting, the counting rate of the liquid sample may increase as much as 100% in the space of 1 hour. This increase could be prevented by the addition of nonradioactive gold chloride or a surface-active wetting agent such as Dreft. Apparently gold chloride in very dilute solutions has a tendency to concentrate at the liquid-air interface. There is no reason to believe this phenomenon is limited to radioactive gold.

Added to the above shortcomings one might reasonably object to the inconveniences of storing liquid samples.

When dried samples are used in conjunction with an end-window tube, most of these problems disappear. Self-absorption is reduced, settling or floating is eliminated, and storing of the sample can be done conveniently. However, to obtain reproducible results it is necessary to present the dried sample to the Geiger tube under conditions of reproducible geometry. This involves a mechanical device that places the sample under the end-window tube in the same orientation every time. It also requires the sample to be dried or precipitated in such a manner that the distribution of radioactivity is uniform or at least reproducible.

These problems are minimized when a wrap-around method is used. Distribution of radioactivity on flexible foil does not affect the counting rate greatly as long as self-absorption does not present a problem. In addition it is possible to increase the efficiency of counting hard beta emitters such as P^{32} by the improved geometry of the sample with respect to the Geiger tube.

Wrap-Around Samples

The authors do not want to imply that the use of wrap-around samples is original with them or even new. It was widely used at the University of California in the early cyclotron days and probably in many other institutions. However, the procedure has been somewhat modified as follows:

Rectangular strips of Reynolds household aluminum foil $2\frac{1}{2} \times 1\frac{3}{4}$ in. are made into cups by folding the edges manually or by use of the sample press in Fig. 1. This cup is placed on a thermostatically controlled hot plate. Radioactive samples in aqueous solutions are added to the aluminum cup in toto or dropwise. When the radioactivity is present in alcohol or ether, the sample is always added dropwise. To prevent creeping of the non-aqueous solvents, a piece of filter paper $1\frac{3}{4} \times 1\frac{1}{2}$ in. is placed in the cup before addition of the radioactive solvent. When the radioactivity is present in dilute sodium hydroxide or hydrochloric acid, lead foil is used instead of aluminum foil.

When the sample is thoroughly dry, the edges are unfolded and the sample is covered with 3-in.-wide Scotch tape. After cutting the corners the tape is folded over the edges to prevent escape of the radioactive material.

The sample is now ready for counting. When counting is done manually it is put around the Victoreen 1B85 Geiger tube and held in place with a wire test-tube clamp. When the automatic sample changer is used the sample is flattened and curved in the roller shown in Fig. 1. The samples are then put into the wells of the turntable (Fig. 2) so that the inside bore of all samples is reproducible within $\frac{1}{64}$ in.

Automatic Sample Changer

There are 41 sample wells around the outer diameter of the turntable, as shown in Fig. 2. These wells have beveled openings, are equally spaced, and are separated by $\frac{3}{16}$ in. of steel shielding plus the plastic walls of the wells. This proves to be adequate shielding for counting energetic beta emitters since normal backgrounds can be recorded between highly active beta samples. For counting gamma-emitting samples, one might replace every other sample well with a lead shield. In this case stopping switch E would have to be actuated by tabs at every other steel separator.

The Geiger tube lifts from one sample and dips into the next. Although only dry counting has been done in our laboratory, apparently the system would meet the requirements of a dipping counter for liquid samples. A dipping Geiger tube used in this manner would have to be silicone coated so that no contaminant remained on the tube to be carried over from sample to sample.

The automatic sample changer is used in conjunction with a Tracerlab Autoscaler. The scaler is set on automatic for counting a preset number of counts, and the time for each sample is printed on a Simplex printing timer. The printing timer was removed from its original housing and incorporated in the automatic sample changer cabinet. Paper tape of the printing timer can be seen on the right of the cabinet in Fig. 2.

At the beginning of each new counting cycle, relay K_1 (Fig. 3) of the scaler is energized. This closes one set of contacts to start the timing cycle of the printing timer and opens another set of contacts to de-energize the mechanical changer circuits.

When the preset count of the scaler is reached, relay K_1 is de-energized. This stops the printing timer, energizes thermal delay relay R_4, and through microswitch B, which is normally closed, energizes impulse relay R_2. Relay R_2 closes the circuit for by-passing switch A. By the circuit parallel to switch A the Geiger-tube lifting motor and its brake-release solenoid LB are energized.

During lifting of the Geiger tube out of one sample and dipping into another, delay relay R_4 closes and energizes relay R_1. The delay relay limits the time of energizing the print circuit to

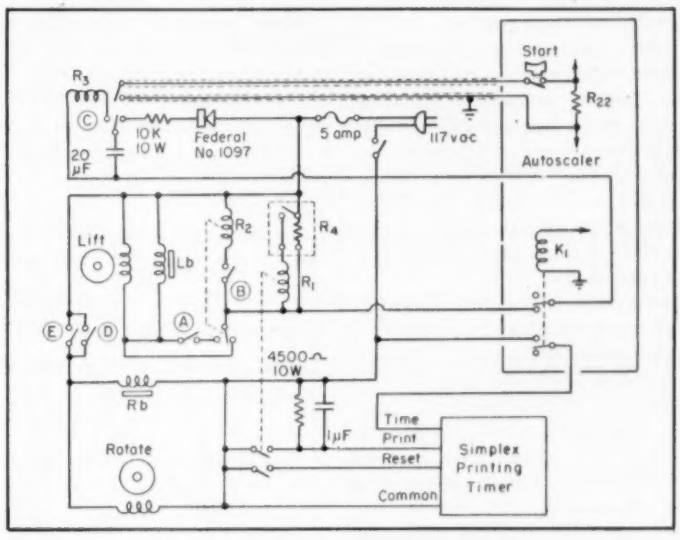


FIG. 3. Schematic circuit diagram of automatic sample changer

a minimum and permits a positive stop of the printing time wheels. Thus, a good clear impression is printed. Energizing relay R_1 closes circuits that cause the printing timer to print and reset.

Microswitches *A*, *B*, *C*, and *D* of Fig. 3 are all cam-operated from the shaft of the back-geared lift motor, which makes one revolution during a sample change. Micro switch *E* is opened by the sample separators of the turntable each time a new sample comes into position. During the sample change cycle switch *B* is momentarily opened by a cam to reset relay R_2 to the circuit of switch *A*, which is normally closed. Switch *D* is momentarily closed by its cam to energize the rotation motor and rotation brake *RB*, even though switch *E* is held open by the preceding stopping action.

As the cycle is completed, switch *A* is opened to de-energize and brake the lifting motor. At the same time switch *C* discharges a 20-μf capacitor through the coil of a 5,000-ohm plate circuit relay R_3. Relay R_3 holds open the circuit across resistor R_{22} of the Autoscaler long enough to reset Autoscaler relay K_1, and the counting of a new sample begins.

The turntable is powered by a back-geared motor (*M*, Fig. 2) mounted to provide friction drive at the table rim. The rotation brake *RB* consists of a solenoid that, when de-energized, positively locks and positions the sample separator partitions. This insures exact alignment of each sample well under the Geiger tube. Should the Geiger tube meet excessive resistance due to a wrinkled or improperly prepared sample, the vertical lift arm disengages from the horizontal arm and opens the main power line by falling against jam switch *S* (Fig. 2). By a simple disengagement of the rotation motor and the rotation brake the operator can quickly position any sample under the Geiger tube for starting a new counting sequence.

The automatic sample changer described was designed for a particularly efficient method of counting dry wrap-around samples of P^{32}. Some possible uses have been suggested that should be of interest to those using other counting techniques.

* * *

The authors wish to acknowledge the assistance of Gardner Ruffin and W. E. Romans, Research Assistants, and F. B. Larson in the construction of this equipment. Development of the equipment was aided by funds from The American Cancer Society and the Life Insurance Medical Research Fund.

Radioactive Contamination Sampling by Smears and Adhesive Disks

By **EUGENE V. BARRY** and **LEONARD R. SOLON**
Health and Safety Division, New York Operations Office
U. S. Atomic Energy Commission, New York, New York

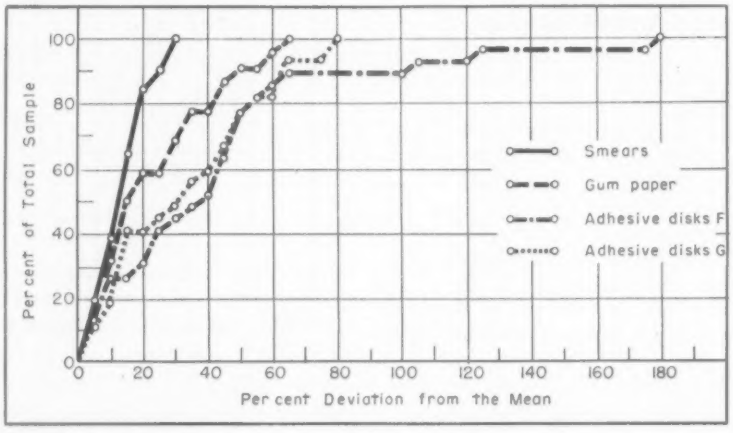
Plants processing radioactive materials and laboratories utilizing radioisotopes detect and measure removable surface contamination by means of various smear and adhesive samples that are then counted with an appropriate instrument for measuring radioactivity. Two primary questions arise in the use of such techniques:

What constitutes removable contamination? The radioactivity hazard is dependent upon the ease with which contamination can be freed from the surface. This varies from surface dust to radioactive materials included in glazes as coloring.

How reproducible is a particular sampling technique? An exploratory investigation of two techniques in current use here for contamination sampling sheds some light on this question; results are reported here.

Technique

Both filter-paper smears and adhesive samples were tested. Uranium metal plates with a normal coating of removable oxide were chosen as a good approximation to a uniformly contaminated surface. These plates were 0.025-in. thick and 3¼ to 3½ in. in width. Consistent results were ob-



FREQUENCY DISTRIBUTION of observations from the mean for contamination sampling methods

Results of Contamination Sampling Tests

Sampling technique	*Plate*	*No. samples* N	*Average (cpm)* $\bar{x}$	*Variance* σ^2	*Standard deviation* σ	$\frac{\sigma}{\bar{x}}$	$\frac{\sigma}{\bar{x}^2}$
Smear	A	6	365	2,446	49	0.135	0.018
Smear	B	4	399	7,017	84	0.210	0.044
Smear	C	7	667	8,117	90	0.135	0.018
Smear	D	3	212	2,086	46	0.215	0.046
Smear	E	11	624	10,697	103	0.166	0.027
Adhesive tape	F	27	204	14,136	119	0.582	0.338
Adhesive tape	G	27	481	38,928	197	0.411	0.169
Adhesive paper	H	22	148	1,975	44	0.299	0.090

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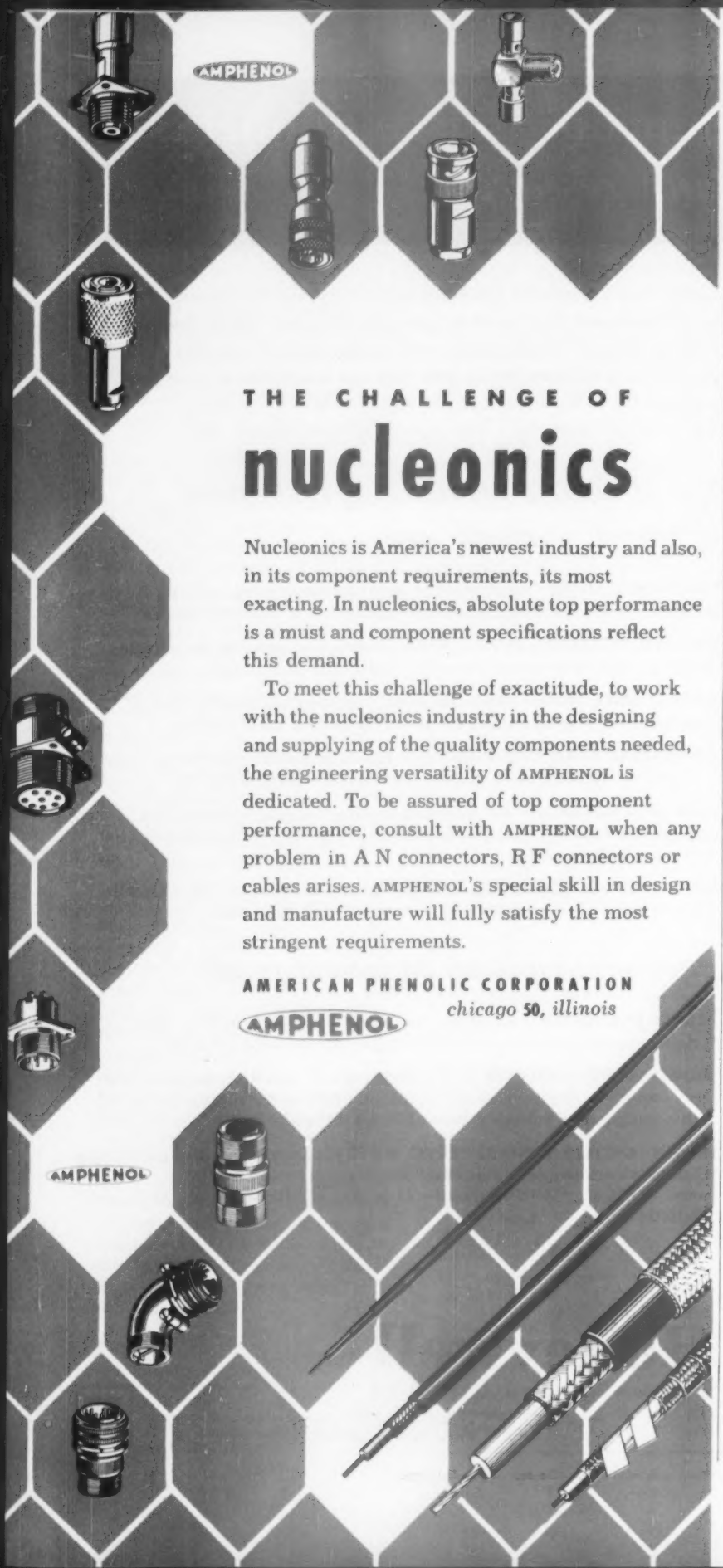
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tained with any one plate, but wide variations in means between plates indicated that the amount of removable material is a fairly sensitive function of the past history of the plates, including manner and place of storage. Filter-paper smears were all taken by one person using 1⅛-in. diameter disks of Whatman 41 paper centered at the bottom of a No. 5½ rubber stopper. The disk was rubbed back and forth across the width of the uranium plate 10 times.

Two types of adhesive disks 1⅛ in. in diameter were used: gummed paper (Simon Adhesive Co. No. 2) and gummed tape (Minnesota Mining and Manufacturing Co. No. 216). Samples were taken by putting the adhesive down on the uranium plate and pressing on the back of the disks with a rubber stopper.

Smears and adhesive samples were placed in a plastic holder for counting with a scintillation detector using a zinc sulfide phosphor.* Count-controlled operation was used for each sample, a total of 640 counts terminating the measurement.

Results

The table on page 60 gives the results of sampling for each uranium plate and method of sampling. Statistical quantities are also given. Apparent differences between the normalized variances were found to be insignificant for the smeared plates.

The illustration on this page shows the frequency distribution of observations from the mean for each of the sampling procedures. The effect of the two uranium plates *F* and *G* on the normalized variances is possibly significant, so results for these sets of samples have been kept separate.

Conclusions

Smears give remarkably reproducible results on surfaces of uniform contamination when the smear procedure is carefully duplicated. All 31 samples fell within 30% of the mean.

Adhesive sampling methods were not nearly as reproducible. However, two reservations should be made. All measurements were made on a smooth surface; on rougher surfaces the adhesive might yield the more reproducible results. Also, the adhesives were adaptations of commercial products and were not designed for radioactive contamination sampling.

* R. T. Graveson, H. D. LeVine, H. J. DiGiovanni, NYO-1523 (1950).

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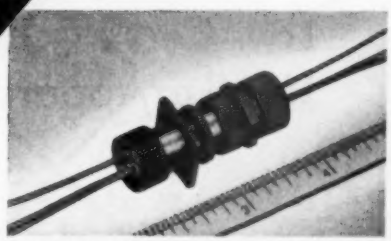

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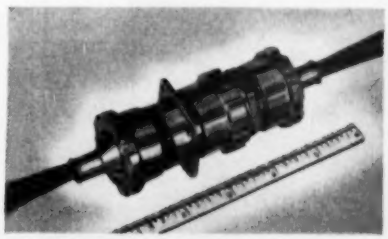

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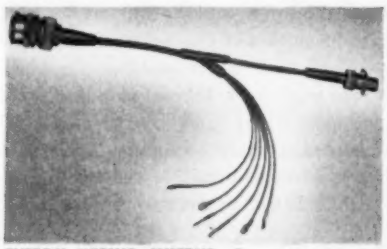

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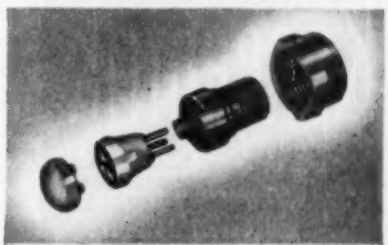

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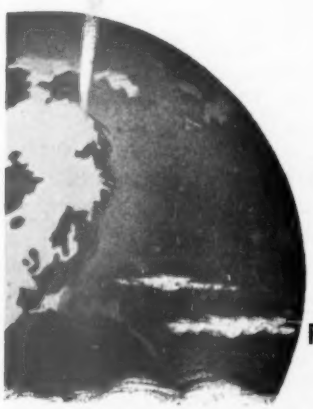

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FIG. 1. Shielded counter for thyroid uptake studies is supported by over-bed hospital table that makes positioning easy

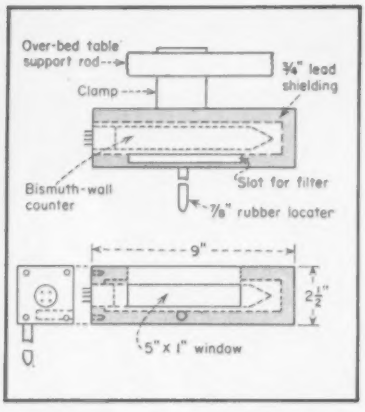

FIG. 2. Construction of shielded housing for counter

Low-Cost I^{131} Thyroid Uptake Apparatus

By EDWARD SIEGEL
Montefiore Hospital
New York, New York

An easily constructed, inexpensive unit for thyroid uptake studies utilizes an overbed hospital table to support a shielded bismuth-wall gamma counter. The unit is used in establishing the functional level of the thyroid gland.

Since only tracer doses of I^{131} are administered for this procedure, and the fraction concentrated in the thyroid counted 24 hr later, accurate counting requires high sensitivity. Shielding must be adequate to exclude counts emanating from extrathyroidal sources such as the bladder, but the counter must receive contributions from the entire, extended and inhomogeneous thyroid region. It is desirable to make the set-up insensitive to small changes in positioning. Finally, the position of the counter housing should be readily adjustable to permit assay of the body "background," or circulating radioiodine level.

Designed for use with ambulatory patients, the unit shown in Figs. 1 and 2 meets these requirements. The shield follows Marinelli's basic lead-shielded housing in design. It encloses a Radiation Counter Laboratories Mark 1 Model 12 bismuth-wall gamma counter. Satisfactory support for the shield is provided by the over-bed hospital table that also makes positioning very simple.

The shielded tube in its counting position is about 7 in. above the skin and has a sensitivity of about 100 cpm/μc of I^{131}. Administration of 15–20 μc of I^{131} provides an adequate counting rate to yield a statistically significant determination in only a few minutes of counting. When reduced sensitivity is desired, the $\frac{1}{2}$-in. lead filter is interposed between the counter and port, giving a $\frac{1}{20}$ attenuation. Patients can thus be assayed after receiving a therapeutic dose.

The total cost of the unit was $275, of which $150 was for construction of the shield housing, $50 for the over-bed table, and $75 for the G-M tube.

* * *

This apparatus was designed for Dr. S. M. Seidlin, New York, N. Y., and is in use in his office. The baffle on the original unit (Fig. 1) is adapted from the basic design of R. S. Yalow, Veterans Administration Hospital, Kingsbridge, N. Y.

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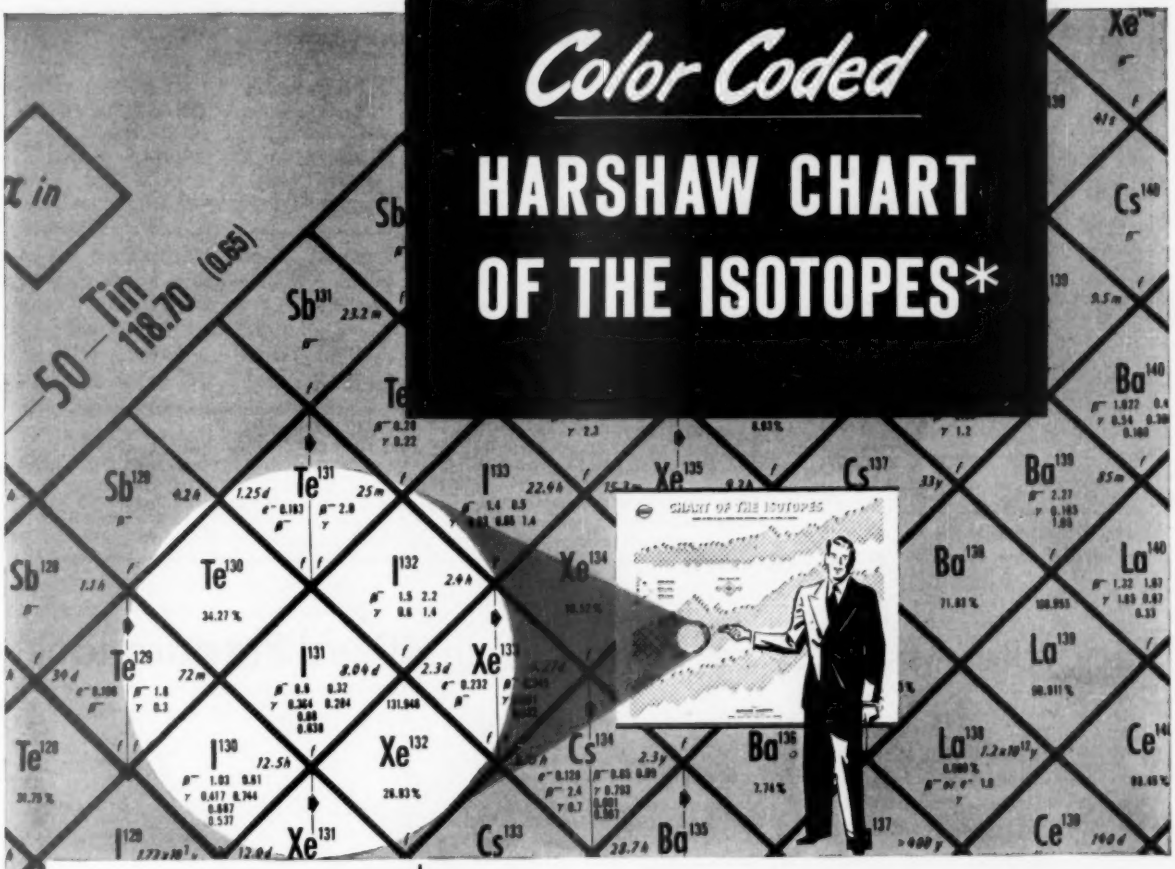

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- **Artificially Radioactive Isotopes Coded by Half-life in 6 Colors**
- **Radioisotope half-life**
- **Modes of Radioactive Decay**
- **Radiation Energies in Million electron volts**
- **Isomeric States**
- **Isomeric Transitions**
- **Nuclear Transmutation Code**
- **Percentage Abundance of Naturally Occurring Isotopes**
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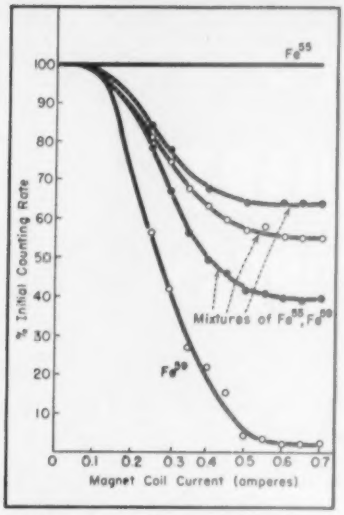

EFFECT of increasing magnetic field upon the counting rate of various samples

Effect of Magnetic Field on Counting Rate of Samples of Fe^{59} and Fe^{55}*

"Units"†		*Counting rate (cpm); magnet*		*Computed rate (cpm) due to:*		*Error in determination (%)*‡	
Fe^{59}	Fe^{55}	*Off* (R)	*On* (R′)	Fe^{59} $\frac{R - R'}{1 - a}$	Fe^{55} $\frac{R' - aR}{1 - a}$	Fe^{59}	Fe^{55}
1	0	650	10.6	...	...	...	...
0	1	975	977	...	...	...	...
1	1	1,641	968	683	948	+5	−3
0.5	1	1,276	974	307	974	−5	0
0.1	1.5	1,575	1,479	100	1,476	+54§	+1
1	0.05	709	59	660	49	+1.5	+0.5

* Based on 6,400 counts collected. Samples were run in duplicate and averaged. a is given by R'/R for the first sample (Fe^{59} only) (10.6/650 = 0.016).

† "Units" are amounts of Fe^{59} and Fe^{55} that yield 650 cpm and 975 cpm respectively.

‡ In addition to counting errors, this includes variation in sample preparation (pipetting, plating, etc.).

§ This large error is primarily due to counting statistics. A very large number of counts must be collected to assess small relative amounts of Fe^{59} accurately since a small difference in counting rates determines the computed value.

Separate Assay of Fe^{55} and Fe^{59} in Mixed Preparations

By W. B. STEWART and H. H. ROSSI

Departments of Pathology and Radiology, College of Physicians and Surgeons
Columbia University, New York, New York

Two radioisotopes of iron, Fe^{55} and Fe^{59}, are of practical use in biological research. While most of the recent work on iron metabolism has been done with the aid of Fe^{59}, there are many experiments in which simultaneous use of both isotopes is of value. For example, populations of erythrocytes of two different ages can be studied simultaneously (*1*, *2*), and corrections for diversion of iron from hemoglobin synthesis can be made in studying iron absorption (*3*).

Fe^{59} has a half-life of 47.5 days and emits beta spectra of 0.46 Mev and 0.26 Mev maximum energy as well as gamma quanta of 1.1 Mev and 1.3 Mev. Fe^{55} has a half-life of 2.9 years and decays by *K*-capture, which results in X-radiation with a maximum energy of 5.6 kev.

Peacock *et al.* (*4*) have devised a method of discriminating against either isotope by employing G-M tubes of different characteristics. Fe^{59} can be preferentially detected by means of a helium-filled tube with a thin mica window. An argon-filled tube with a beryllium window serves as a detector that is mainly sensitive to Fe^{55}. As far as we are aware, this is the only selective method reported in the literature.

The approach in the method reported here has been to suppress the beta radiation from Fe^{59} by means of a magnetic field. When such a field is applied at right-angles to the trajectory between the sample and the counter, the electrons may be deflected through 180 degrees and prevented from reaching the counter. Thus Fe^{59} can affect the counter only by means of its gamma radiation, which is detected with low efficiency. On the other hand the X-radiation from Fe^{55} is unaffected by the magnetic field and is detected equally well with or without the field.

A soft iron yoke was constructed for two coils that were available. The pole pieces are truncated cones with 1.5-in. minor diameters. The maximum field between them is about 1,850 gauss. Complete suppression of all beta radiation requires a separation of twice the radius of curvature of the particles of maximum energy. For these electrons (0.46 Mev), $H\rho$ is about 3,000, and a sample-to-counter distance of about 3.3 cm was indicated. This figure is a lower limit since air scattering tends to increase the penetration of the radiation. In practice, a separation of about 4 cm is adequate for total suppression of the beta radiation from Fe^{59}.

The figure on this page shows the effect of increasing magnetic field on the counting rate obtained with pure samples of Fe^{55} and Fe^{59} and three mixed preparations. The radioactive material was electroplated onto copper disks by the method of Hahn (*5*). The counter employed is a thin-window type (Nuclear D 34) that has an iron ring near the sensitive end. A different type of counter without such shielding (Tracerlab TGC-3) exhibited a drop in sensitivity to Fe^{55} by about 10% at maximum strength of the magnetic field. It is believed that the magnetic field interfered with the operation of this tube.

Under the conditions of the experiment the counting rate of the pure Fe^{55} sample was unaffected by the magnetic field, while the sensitivity

to Fe^{59} was reduced to about 3%. This residual response was due to the gamma radiation. While such a high degree of differentiation is not necessary in many experiments, it has the advantage that errors in greatly unequal mixtures are minimized. Furthermore, if the intensity of the magnetic field exceeds a critical value, the counting rate becomes independent of field strength.

The counting efficiency in this arrangement is somewhat low (about 1% for Fe^{59} and 0.05% for Fe^{55}) but is adequate for the experiments in progress. It could obviously be improved substantially by the employment of a stronger magnetic field, which would permit a shorter sample-to-tube distance.

A correction can be made for the small residual sensitivity to Fe^{59} at maximum magnetic field strength. If F is the counting rate due to Fe^{59}, F' the counting rate due to Fe^{55}, R the counting rate with no magnetic field present, R' the counting rate with the magnetic field present, and a the factor by which the magnetic field reduces the counting rate of Fe^{59}, then

$$F + F' = R$$
$$aF + F' = R'$$

Hence

$$F = \frac{R - R'}{1 - a}$$
$$F' = \frac{R' - aR}{1 - a}$$

These corrections have been applied to the figures given in the table on this page that shows data obtained with various mixtures of the two isotopes.

The advantage of this method is that results may be obtained with a single Geiger counter by determining counting rates with and without a magnetic field. These measurements can be made with a single assembly under fixed geometrical conditions.

The method could be employed to differentiate between any other beta-emitting isotopes and X-ray emitters. It might be of some help in distinguishing between beta emitters of greatly different disintegration energies.

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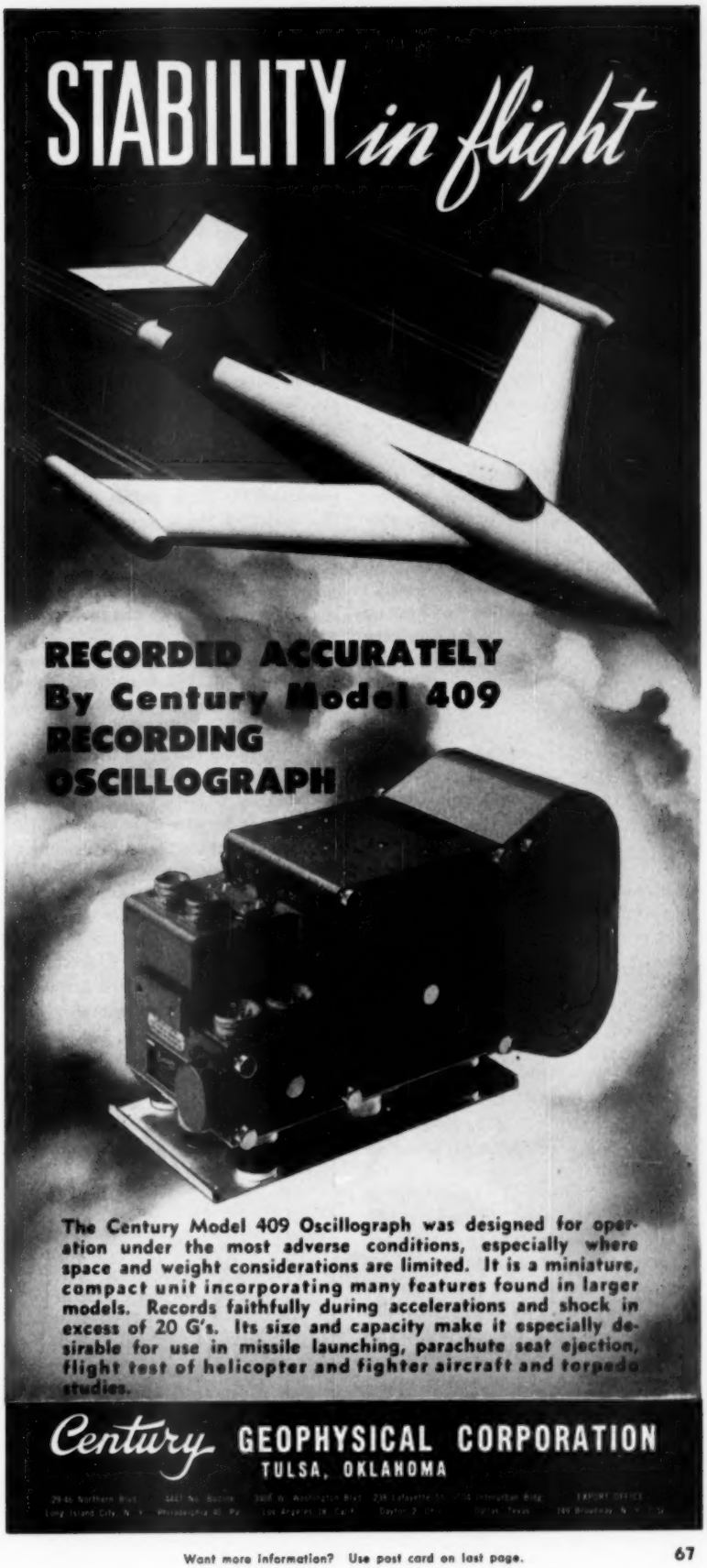
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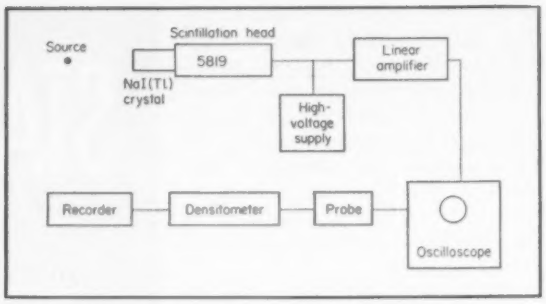

FIG. 1. Block diagram of γ-ray scintillation spectrometer

This method for determining the pulse energy distribution produced by γ-rays incident on a NaI (Tl) crystal offers several advantages:

1. A γ-ray spectrometer that is rapid, simple, accurate and versatile.

2. When compared with other systems, such as the elaborate multichannel electronic analyzers, there is no sacrifice in either resolution or speed.

3. Although no increased amount of activity is required, the time necessary to obtain a complete pulse energy spectrum is short compared with that needed by other simple (single channel) scintillation spectrometers.

Problems in medical physics demand a fast γ-ray spectrometer. The amount of absorption and scattering occurring in sources of finite size within a body can be determined only from an analysis of the emitted γ-ray spectrum. If such sources contain mixtures of two radioisotopes emitting γ-rays of different energies, the ratio of their activity can be determined with the spectrometer. Localization of sources within a scattering medium can be improved by discriminating between primary and scattered radiation.

A schematic diagram of the spectrometer system is shown in Fig. 1. The triggered pulses are displayed on the screen of a Tektronix 513D oscilloscope, using a sweep time of 0.1 μsec/cm. Instead of employing a photographic method* for pulse-height analysis, the light intensity distribution on the screen is measured directly by a photoelectric cell. For this purpose, a densitometer built by the W. M. Welch Scientific Company is employed. This densitometer is a stable instrument that has a sensitivity adjustable over a wide range. Its output current is recorded by a Weston recording potentiometer. An advantage is that the final result is immediately available.

The pulse spectrum is displayed on the oscilloscope screen so that all pulses have a horizontal section of at least 1 cm in width at the vertical center axis of the screen. The light produced by those pulses present at the center of the screen is channeled into the photoelectric cell through a Lucite rod. The light can enter the latter only through a 0.3 × 1.0 cm slit that is in contact with the oscilloscope screen.

For recording the spectral pulse distribution, the spectrum is moved slowly and at a uniform rate across the oscilloscope screen. The potentiometer controlling the vertical position of the Tektronix oscilloscope has been replaced by a 10-turn Helipot driven by a 1-rpm synchronous motor. The gain of the amplifier is adjusted so that the pulse spectrum is spread out beyond the sensitive part of the oscilloscope screen. Therefore, only a fraction of the complete pulse spectrum is visible at one time. If the motor-driven vertical positioning control is activated, the complete spectrum moves slowly across the screen. Since the amount of light produced by only those pulses passing through the center of the screen is measured, variations in screen sensitivity do not show up.

Figure 2 shows the pulse spectrum of a Hg^{203} source. Both the 279-kev γ-rays and the thallium-K X-rays produce high-intensity photoelectric peaks. The solid curve is plotted from a recording using the spectrometer system shown in Fig. 1; the points are measurements taken with a single-channel pulse-height analyzer (2-volt window) and scaler. There is good agreement between the two methods. However, not all the low-energy pulses show up on the screen, since the probability of triggering is due mainly to those pulses of higher energy. The recorded spectrum is not affected by an increase in source strength or a change of the light intensity of the oscilloscope, if the densitometer sensitivity is adjusted accordingly.

Figure 3 is an actual recording of two I^{131} energy spectra. The source was 10 μc of I^{131} in aqueous solution at 10 cm distance from the large NaI (Tl) crystal (1¾ in. diameter, 2 in. height). The total counting rate was approximately 250,000 cpm. Besides the

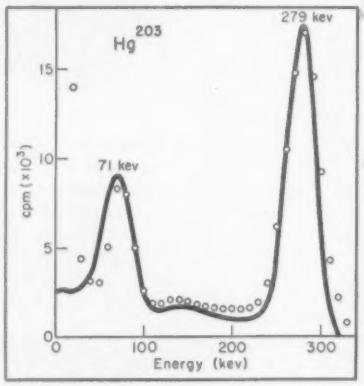

FIG. 2. Hg^{203} pulse energy spectrum. Solid curve taken from a recording obtained in ten minutes. Points determined with single-channel pulse-height analyzer

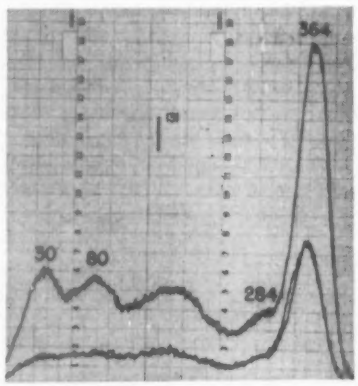

FIG. 3. Double recording of I^{131} pulse energy spectra. Low-intensity spectrum obtained with a 2.5-mm Pb filter between source and NaI(Tl) crystal

* W Bernstein, R. L. Chase, A. W. Schardt, *Rev. Sci. Instr.* **24**, 437 (1953).

well-known γ-ray lines resulting from the decay of I^{131}, X-rays of about 30 kev are also apparent.

The second spectrum with reduced intensity was obtained with a 2.5-mm lead absorber placed between source and crystal but otherwise identical conditions. The fluorescent radiation emitted from the lead absorber was filtered by a tin-aluminum combination added to the lead absorber. Two recordings of each spectrum have been superimposed. They agree with each other within the expected statistical variations.

The γ-ray spectrometer described has been assembled from commercially available equipment. It has proved to be a very reliable, stable instrument that can easily be maintained. The recorded pulse distribution is obtained with good accuracy in a short time.

* * *

The technical assistance of Mr. L. Sarkes is gratefully acknowledged. This investigation was supported by the Veterans Administration and is published with the approval of the Chief Medical Director. The statements and conclusions published by the author are a result of his own study and do not necessarily reflect the opinion or policy of the Veterans Administration.

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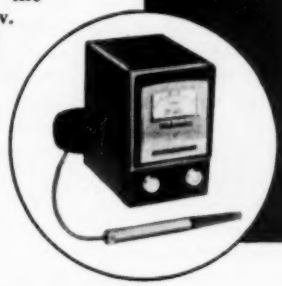

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BOOKS

Refractory Hard Metals

By PAUL SCHWARZKOPF and RICHARD KIEFFER (Macmillan Co., New York, 1953, xi + 447 pages, 91 illustrations, $10.00). Reviewed by HENRY H. HAUSNER, Atomic Energy Div., Sylvania Electric Products, Inc., Bayside, New York.

The term "refractory hard metals" as used in this book refers to high-temperature-withstanding materials, such as the borides, carbides, nitrides, and silicides of the transition metals. These substances are mostly used in cemented state, i.e., in combination with binder metals such as cobalt. In this book, the term "hard metals" refers to the binder-free substances as well as to the cemented materials. Another reader may have some doubt as to a tungsten boride or a uranium carbide being considered a metal. The authors, however, believe that the physical properties of these materials, especially their electric conductivity, indicate clearly a metallic bonding of the binder-free substances, and the term "metal" is therefore justified.

The book deals with the structure, preparation, and properties of these hard metals and with their application as constituents of the so-called "super-alloys" for high-temperature application. The materials described in this book are of utmost importance for the development of gas turbines, jet engines, and rockets, and also for nuclear reactors and other nuclear applications. The engineers of today think frequently in terms of temperatures of 1,000° C, or even higher, and iron-base alloys can no longer be considered for many of these new applications.

Both authors are internationally known pioneers in the field of powder metallurgy, especially in the development of hard metals, and there are hardly any other investigators so well suited to write a book on this subject. Its content is mainly based on the experience gained in Dr. Schwarzkopf's laboratories and production plants in the United States, Austria, and England. The only other existing book concerned exclusively with hard metals is Karl Becker's "Hochsohmelzends Hartstoffe," Verlag Chemie, Berlin, which was published twenty years ago and which has not been translated into English. The book under discussion is, therefore, the first book in the English language on this important branch of powder metallurgy and provides an up-to-date presentation of the history and current trends of hard metal development. It is based on a critical evaluation of the literature, supplemented extensively by previously unpublished material originating from the experience of the authors and their collaborators in two continents.

The forty-four chapters of the book are divided into six groups. The first two chapters deal with the crystal and electronic structure of refractory hard metals. The excellent presentation of this material is based mainly on the work by Hagg, Kiessling, Koster, Pauling, and Ubbelohde.

The largest group of chapters is devoted to the carbides, their preparation and purification, especially the carbonization of metal and oxide powders with solid carbon or with carbonaceous gases. For the readers of NUCLEONICS, it will be of special interest that one chapter in this group is devoted exclusively to the carbides of the metals of the actinide series, such as thorium, uranium, and plutonium carbides, although these metals cannot be classified as transition metals. Similar chapters within three other groups in the book deal with the borides, nitrides, and silicides of thorium, uranium, plutonium, and other metals of interest for nuclear application. The behavior of these materials at elevated temperatures is described.

A special chapter is devoted to the multicarbide systems and the solid solubility between carbides. The important nitride-carbide systems, as well as the boride-carbide and the boride-nitride systems, are extensively described and discussed. The reviewer was especially attracted to a chapter on molybdenum-silicide, a material which may play an important role as a heating element for high temperature furnaces; its preparation and physical properties are described in detail and many references are given.

The authors believe that an engineering evaluation of the carbide, boride, and silicide materials proposed for high-temperature application must be based on a comparison with the performance not only of super-alloys, but also of some ceramic materials and sulfides developed for high-temperature service. The last two chapters of the book include a discussion of super-alloys, special ceramics and sulfides. Since the authors endeavored to present an objective picture of the present status of high-temperature materials, these chapters appear to represent the most up-to-date discussion dealing with their availability and the problems involved in their production and their use in applications requiring mechanical, as well as chemical stability at extremely high temperatures.

The final chapters of the book deal with the oxides of beryllium, zirconium, and thorium, their creep up to 1,000° C and their other physical properties up to 1,500° C.

A carefully prepared author and subject index facilitates the study of the book, which is of greatest value for every scientist, metallurgist, and mechanical and other engineer working in the field of hard metals and high-temperature-resisting materials. It is the first complete reference book in the field.

In the introduction, the authors state that they are preparing a second volume which will deal especially with the production and application of the cemented hard metals in general, and chiefly with cemented carbides. Readers of the first volume described here will look forward with interest to the second volume.

BOOKS RECEIVED

Scintillation Counters, by J. B. Birks, McGraw-Hill Book Co., Inc., New York (or Pergamon Press Ltd., London), 1953, vii + 148 pages, $4.50. (*To be reviewed.*)

Mathematical Aspects of the Quantum Theory of Fields, by K. O. Friedrichs, Interscience Publishers, Inc., New York (or Interscience Publishers Ltd., London), 1953, viii + 272 pages, $5.00. This book deals with linear problems in the quantum theory of fields.

Atomic Medicine, 2nd edit., edited by Charles F. Behrens, Williams & Wilkens Co., Baltimore, 1953, xiv + 632 pages, $11.00. (*To be reviewed.*)

Relativity and Reality, by E. G. Barter, The Philosophical Library, Inc., New York, 1953, xi + 131 pages, $4.75. The subtitle is "A Re-interpretation of Anomalies Appearing in the Theories of Relativity." "This is an attempt to bring the theory of relativity into complete accordance with our common-

sense ideas of space and time, and thus, so to speak, straighten out the world again." [It's about time.]

Fatigue of Metals, by R. Cazaud (translated by A. J. Fenner), Philosophical Library, Inc., New York, 1953, xiv + 334, $12.50. Essentially a translation of the 1948 edition, this book also discusses research done since that time. The subjects covered include theory of metal fatigue, fatigue testing machines, fatigue limits of metals and alloys (with tables), and design of structural joints and machine components.

Advances in Biological and Medical Physics, vol. 3, edited by John H. Lawrence and C. A. Tobias, Academic Press, Inc., New York, 1953, x + 368 pages, $8.00. Of special interest in the nuclear field are the chapters, *The Dosimetry of Artificial Radioactive Isotopes*, by W. V. Mayneord and W. K. Sinclair, *The Kinetics of Biological Processes—Special Problems Connected with the Use of Tracers*, by A. K. Solomon, and *Primary Ionization as a Test of Molecular Organization*, by Ernest Pollard. The first chapter concerns the practical application of dosimetry to the patient. The second chapter treats the following special cases: isotope dilution, uptake from a constant source, closed two-compartment system, closed two-compartment system with leakage from the inner compartment, open two-compartment system—the question of precursors, open two-compartment system with transport coefficients dependent upon the age of the molecule, and diffusion or flow out of a compartment. The last chapter discusses the measurement of sensitive volumes, e.g., molecular volumes or weights, by using bioassay methods together with the action of ionizing radiation.

ALSO OF NOTE

Review of Current Research and Directory of Member Institutions (Engineering College Research Council, American Society for Engineering Education), 1953, xii + 330, $2.50. The 103 institutions covered include all major engineering schools in the United States. The index lists approximately 5,000 research project subjects. For each project, administrative officers, policies that govern projects, personnel, annual expenditures, sources of income, and special conferences and short courses of interest to research workers are given.

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NEWS

Power Study Groups Regroup to Form Team Aiming at Power-Only Reactor; Sixth Group Seeks AEC Approval

Three of the five industrial teams studying the feasibility of atomic power have completed a major realignment of their study groups. In addition, a sixth group new to the field has applied for government permission to initiate atomic power studies. Both moves, of course, are subject to approval of the Atomic Energy Commission. There seems little doubt, however, of AEC's okay.

Prospective new entrants in the field are Duquesne Light Co., Pittsburgh Utility Firm, and Walter Kidde Nuclear Laboratories, Inc.

Reshuffle of the veteran teams will bring together Union Electric Co. of Missouri, Commonwealth Edison Co., Pacific Gas & Electric Co., Bechtel Corp., and American Gas & Electric Co. Chairman of the new group is J. W. McAfee, president of Union Electric, who told NUCLEONICS that: "Dual-purpose reactors do not look as bright to us now as they once did. We want to devote ourselves more exclusively to straight power. And our interest is in full-scale power reactors rather than small plants."

First step of the proposal submitted to AEC is "the selection of a reactor and preliminary design work." Comwealth Edison, which had requested AEC authorization to build a power reactor—reportedly cooled by high-pressure water—has withdrawn this application, at least for the present. This is to give the new group an opportunity to consider this and other types of reactors.

Public Service Co. of Northern Illinois, formerly associated with Commonwealth Edison in its atomic studies, has merged with that company. Thus, Public Service personnel will continue to be available for cooperation with the new study group.

Union Electric's former partner, Monsanto Chemical Co., has asked AEC permission to go ahead with its atomic power work alone. Monsanto has repeatedly discussed with AEC officials the construction of a reactor for the production of commercial electric power and plutonium for sale to the government. Monsanto's president, Charles A. Thomas, has long advocated such dual-purpose plants.

The new five-company group fits the pattern of mass corporate backing followed by two other study teams. Dow Chemical Co. and Detroit Edison Co. are now associated with 25 other firms in their feasibility studies. Pioneer Service and Engineering Co. and Foster Wheeler Corp. have the support of 6 utilities.

The Dow-Detroit Edison group already is conducting advanced studies of the feasibility of a breeder reactor. The Pioneer Service-Foster Wheeler team began its studies only a few months ago.

Phillips Takes Over Chemical Plant in Idaho

Operation of the chemical processing plant at the National Reactor Testing Station in Idaho has been taken over from American Cyanamid Company by Phillips Petroleum Company. This plant has been in operation since last February.

Phillips also runs the materials testing reactor in Idaho.

Schedule Symposium on Scintillation Counters

A Scintillation Counter Symposium will be held on January 26 and 27, 1954 in Washington, D. C., under the sponsorship of AIEE, IRE, AEC and NBS. Four sessions will cover energy spectrometry, cosmic-ray and high-energy particle research, and phosphors and photomultipliers.

The program will include a group of invited papers and a limited number of short contributed papers (fifteen minutes). Title and abstract of proposed contributions should be submitted to R. K. Swank, chairman, program committee, Argonne National Laboratory, Box 299, Lemont, Ill., by Dec. 15.

Advance registration is required for attendance. Those planning to attend should forward their application, together with a $2.00 registration fee, to H. O. Wyckoff, chairman, attendance committee, National Bureau of Standards, Washington, 25, D. C.

The program is designed, in so far as possible, to present recent advances in scintillation-counter technology. Design and performance of instruments and components will be considered. Application of the scintillation counter to the solution of both industrial and scientific problems will be discussed.

Chairman of the Scintillation Counter Symposium committee is G. A. Morton, RCA Laboratories, Princeton, N. J.

Army Studies Costs of Small Power Reactors

Preliminary studies of the feasibility of nuclear reactors to supply electric power for remote military bases have been undertaken by the Army. Objective of the studies is to estimate the costs of such power and assess it against orthodox power.

Bases being considered and other details of the study are under heavy secrecy wraps. But the Defense Department for some time has buzzed with speculation over the feasibility of installing atomic power plants at outposts like the Thule, Greenland, Air Base, where fuel supply represents a serious problem. There has been some talk, too, about power reactors at island bases, where fuel supply lines might be interrupted by enemy air or submarine attacks in event of war. Present power costs at these installations range from 1.3 to 6.0 cents per kwh, excluding capital costs (NU, April '53, p. 32).

Only one power reactor of pertinent capacity and design for the Army study is in existence. This is the land-based prototype of a submarine propulsion unit, now undergoing tests at the Atomic Energy Commission's proving grounds near Arco, Idaho. Built by Westinghouse Electric and AEC's Argonne National Laboratory, this unit reportedly has a useful capacity of upwards of 12,000 kw. Some authorities estimate that a similar reactor now can be built to produce power at costs ranging from three to six cents per kwh.

Curtis W. McGraw Dies; Publishing Company Head

Curtis W. McGraw, president and chairman of the board of McGraw-Hill Publishing Co., of which NUCLEONICS is a part, died unexpectedly on Sept. 10. He was 57 years old.

Mr. McGraw, third son of the late James H. McGraw, Sr., who founded the publishing concern, had been associated with the company since 1920. He was elected president and chairman of the board in February, 1950. His leadership will leave an enduring mark on the pages of this and other McGraw-Hill publications.

Born in Madison, N. J., Mr. McGraw was a graduate of Princeton University. Active in numerous civic affairs, he was a member of the Postmaster General's recently appointed Advisory Board of the Post Office Department. He was a past director and past president of the American Book Publishers' Council.

He was also a director of the First National Bank of Princeton, the Dragon Cement Co., the Princeton University Press of which he was treasurer, the Princeton Hospital of which he was president, and the Magazine Publishers' Association.

Mr. McGraw was a member of the advisory board of the Times Square Branch of the Chemical Bank and Trust Co., a member of the Men's Committee of the Lighthouse, New York Association for the Blind, the Men's Committee of the American Museum of Natural History, the National Association of Manufacturers, the National Society for Crippled Children and Adults, Inc., of which he was a trustee at large, the New Jersey Republican Finance Committee, and of the United States Golf Association.

Hearings Transcript Shows Broad Interest in Effect of Atomic Energy Legislation

A broad cross section of American industry, government, science and the public was represented at the recently concluded hearings on atomic energy legislation conducted by the Joint Congressional Committee on Atomic Energy (see NU, Sept. '53, p. 9, for analysis of hearings). This is brought out in the 649-page transcript of the hearings just released.

A majority of the witnesses and those submitting statements for the record avoided making specific recommendations, either because they preferred not to be pinned down or because there was insufficient time to prepare detailed statements. However, most of the witnesses have indicated that they will prepare very specific proposals by October or November, well in advance of the next group of hearings scheduled for the spring.

Below are several excerpts from statements not available at the time of preparation of NUCLEONICS' September analysis of the hearings:

Manson Benedict, professor of nuclear engineering, MIT: I believe that a useful step in liberalizing security provisions could safely be made by giving properly cleared individuals all relevant technical and economic information needed for intelligent development of nuclear reactors. At the present time, the administrative interpretation of the security provisions of the McMahon Act still results in undesirable compartmentalization of information, particularly the withholding from individuals not employed by the Atomic Energy Commission of information on costs of uranium and other fission materials, and the costs of building and operating Government-owned facilities for processing these materials. Private companies seeking to develop economic atomic power-plants should have all available information on the costs of related Government-controlled operations.

I should like to see a number of pilot-plant investigations made on promising types of nuclear reactors. The homogeneous reactor experiment, which is being conducted at the Oak Ridge National Laboratory, and the experimental breeder reactor, which is being tested by the Argonne National Laboratory, are examples of the type of undertaking which should be multiplied. There are a half-dozen other types of nuclear reactors for the production of useful power which cannot be evaluated properly at the present time for lack of pilot-plant experience, which would give us badly needed information on the life of equipment, cost of construction and operation, and efficiency of conversion of nuclear fuels to electric power. Private companies should be encouraged to do this pilot-plant work with their own funds. To this end, they should be permitted to take out and use such patents as might result from a privately financed pilot-plant program and to own full-scale atomic power plants which might be built as a result of pilot-plant findings.

John Von Neumann, AEC General Advisory Committee: I would like to argue in favor of shifting the emphasis more toward guaranteed Government purchases of plutonium. I think it unlikely that commitments of this nature should turn out to be undesirable in any foreseeable future, or that any degree of availability of plutonium that we will plausibly achieve can prove to be uneconomical or excessive. The potential uses for this material are more varied than is now generally recognized.

K. S. Pitzer, dean, College of Chemistry, University of California: There should be a general separation of important atomic-weapon information from other presently classified atomic information. At one time there was an overlapping of weapon and reactor information. Without possible involvement in classified information, I cannot explain in detail the reasons, but I am convinced that it is now possible to segregate all really important weapon data from the reactor area. Our security of weapon data would be greatly improved if it were clearly segregated and if security clearances for the weapons area could be handled separately from other areas. The Commission should be given broad authority to handle reactor data as it deems best. I believe that a great body of presently classified reactor data might well be published. However, it should also be possible for

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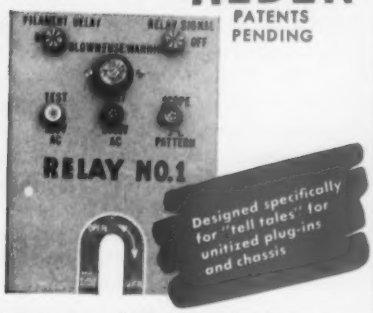

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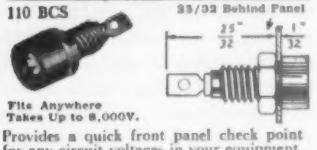

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ELECTRON SCATTERING APPARATUS is being used at Stanford University to study structure of the nucleus. Robert Hofstadter (center) is director of the project; his associates are H. R. Fechter (left) and J. A. McIntyre. Stream of electrons from linear accelerator enters drum-like chamber containing target material through pipe at left. Electrons scattered by target nuclei emerge from long aluminum window, pass into magnetic field for measurement

reactor data to be handled at a low classification without the limitations now associated with atomic energy restricted data.

Homer S. Meyers, president, Radioactive Products, Inc.: While the companies who are interested in the development of economic nuclear power are capably represented in respect to such amendment, I wish to speak in behalf of another segment of this industry which might be considered as a "fringe" or incidental activity to that of nuclear power production.

Specifically, I refer to companies such as Radioactive Products, Inc., who are engaged in the secondary distribution of radioactive isotopes and are engaged in the development of new applications of these materials to various industrial and other problems. These companies are, for the most part, quite new and small but are engaged in solving problems relating to most of the major industries of our country.

There are a number of aspects of the present Atomic Energy Act which seriously hinder the activities of these companies. As an example, I should like to review some of the difficulties which might be encountered by a company such as ours who wishes to evaluate and develop an application of byproduct materials involving extremely high radiation levels such as may be encountered in cold sterilization or the catalysis of a particular chemical reaction.

First, this investigator is unable to acquire information as to the materials that are potentially available from a large-sized operating reactor. Such information is generally available only to the personnel of contractors of the Atomic Energy Commission who have a specific need for this information in connection with that contract. If he is employed by one of the larger companies, on the other hand, his opportunities are much greater since it is quite likely that the large company will have an Atomic Energy Commission contract sufficiently pertinent that he can acquire this exploratory data. Let us suppose, however, that this investigator is able to acquire the necessary information, and determines that his proposed application may be feasible. He would like to purchase a fuel element recently removed from a reactor, or else to have an irradiation of his own uranium sample made in one of the reactors.

It is only by the broadest interpretation of the law (as I understand it) that such transfer is allowable. Were this broad interpretation made, however, and were he to receive the material or services requested, he would be immediately involved in a tremendously complicated and expensive procedure in accounting for every trace of the fissionable material which had passed into his hands. He would, further, be in possession of physical objects and information which would require ex-

treme security measures. He would assume liabilities beyond any ordinary industrial practice for the perpetual safe handling of the materials that came into his possession. His distribution and use of these materials would henceforward be under the control of the Atomic Energy Commission. Certainly a consideration of this succession of difficulties would discourage the initiation of any but the most obvious likely applications.

NICB to Hold Meeting on Atomic Energy in Industry

The second annual conference on "atomic energy in industry" will be held by the National Industrial Conference Board in New York, October 29, 30.

There will be six technical sessions: changing the Atomic Energy Act—pro and con; reactor design and technical developments in new metals and coolants; developing commercial atomic power—analysis of feasibility studies; cutting costs through use of radioisotopes; possible economic impact of atomic energy; and improving product quality with use of radioisotopes.

In addition, luncheon and dinner sessions will feature W. Sterling Cole, chairman Joint Committee on Atomic Energy; David E. Lilienthal and Gordon Dean, former chairmen of AEC; and Sir Christopher Hinton, leading British atomic energy figure.

AEC Not to Build Eureka Power Reactor

The Atomic Energy Commission has decided that "it would not be practical at this time to locate a nuclear power plant in Eureka, Nev." Studies made of other high-power-cost areas in the U. S. brought AEC to the same conclusion.

The Nevada Atomic Power Utilization Committee proposed to AEC last year (NU, Sept. '52, p. 66) that a power reactor be built at Eureka. AEC's report on the Eureka region, just released, points out:

"1. At 10 mills/kwh cost, the demand for power by 1960 would approximate 30,000 kw at 60% load factor.

2. From 10 to 15 mills/kwh the demand would fall off gradually; above 15 mills it would drop rapidly.

3. By construction of a transmission line from contiguous areas such as Twin Falls, Idaho; Summit, Calif.; and Salt

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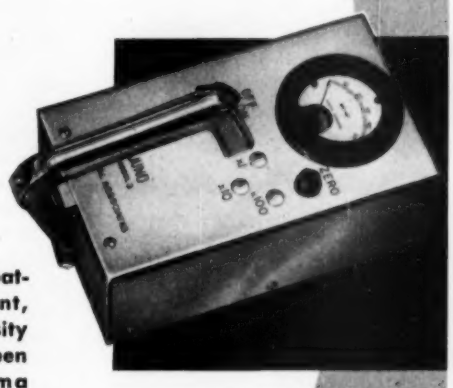

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Lake City, Utah; the cost of power delivered to Eureka is estimated to range from 10 to 13.1 mills/kwh.

4. Power of the required magnitude and load factor would cost from 20 to 25 mills from reactor sources of the small size needed, assuming present technology."

Power Program Unchanged by Russian H, A-Bombs

Prospects for development of industrial electric power from fissionable fuels are undimmed by recent Russian experiments with both fission and thermonuclear weapons.

That's the concensus of informed sources in Washington, both in Congress and in the military and other administrative agencies.

Briefly, here is how official Washington took the news that Russia had conducted thermonuclear tests on August 12 and started experiments with apparently far-advanced explosives of the fission type on August 23.

This was about the kind of progress our military and atomic energy experts had estimated the Russians would make in this field several years ago. The Reds may have been a few weeks, perhaps months, ahead of the timetable estimated for them in Washington, but no more. So both our military and atomic energy programs have been geared to meet just this type of Russian advancement.

Nowhere in Washington is there a demand for expansion of Atomic Energy Commission's production and development programs. True, there probably will be calls next year when Congress convenes for additional spending for air defense as a result of the Russian tests. Any change in the atomic energy program, however, probably will go to military applications rather than to the supply of fissionable materials for that purpose.

Before leaving with several other members of the Joint Congressional Committee for a trip to South Africa, Chairman W. Sterling Cole reaffirmed the Committee's schedule for hearings on proposed amendments of the Atomic Energy (McMahon) Act of 1946. The Committee still plans to get these hearings under way early next year, get amendments designed to promote private development of nuclear power before Congress by April or May.

AEC officials also discounted the possibility that military demands for fissionable materials will increase so sharply as a result of the Russian tests as to make diversion of such material for industrial purposes impossible. They pointed out that the military program for development of nuclear weapons, like AEC's own expansion program, was designed to meet just such eventualities as the now apparent Russian program.

AEC people also emphasized that diversion of materials for industrial nuclear power still is years away, will not be necessary until after a plant is actually built. The first diversions will be comparatively insignificant. Finally, a thriving nuclear power industry eventually will be an additional source of fissionable materials that can be tapped in the event of a military emergency.

These views were shared at many levels in the Defense Department.

IN BRIEF

• **World nucleonics.** On a proposal by Gunnar Randers, director of the Joint Norwegian-Dutch Atomic Energy Establishment, the participants in an international conference on industrial nuclear power unanimously voted in Oslo last month to establish an international nuclear energy association, to be open to scientists of all nations. Organizational details will be worked out by a five-man committee, subject to approval by scientific bodies in the participating countries.

• **Production plant.** A classified production facility will be built by AEC at the Lake Ontario (N. Y.) Ordnance Works at a cost not to exceed $5-million. Singmaster and Breyer, New York City, will be the architect-engineer contractor to design the facilities, and the Hooker Electrochemical Co. of Niagara Falls the operating contractor.

• **Contract extended.** The agreement between AEC and Union Carbide and Carbon Corp. has been modified to include operation of additional facilities at K-33 and Y-12 plants, Oak Ridge, and the gaseous diffusion plant, Paducah, Ky. The terms of the agreement were also extended two years, to June 30, 1957.

NEWSMAKERS

E. P. Lee will head a new department that will test a prototype atomic submarine power plant at Knolls Atomic Power Laboratory. **J. H. Warren** replaces Lee as manager of the manufacturing department reactor section

at Hanford, and **R. O. Mehann** assumes Warren's post as reactor section production superintendent.

Lawrence C. Widdoes has taken a two-year leave of absence from Monsanto Chemical Co. to become project director for the research reactor being built by the University of Michigan.

James W. McRae, vice president of Bell Telephone Laboratories, has succeeded Donald Quarles as president of Sandia Corp.

Three department heads have been appointed by Walter Kidde Nuclear Laboratories: **C. B. Ellis,** research dept., laboratory division; **J. J. Byrnes,** engineering dept., laboratory division; and **Karl Puechl,** theoretical dept., development division.

W. W. Grigorieff, director of the Institute of Science and Technology of the University of Arkansas, has been named chairman of the university relations division of the Oak Ridge Institute of Nuclear Studies to replace **Russell S. Poor.**

Capt. Robert A. Hinners, USN, has been appointed director of the Navy's Radiological Defense Laboratory in San Francisco. He was previously with the Armed Forces Special Weapons Project in Washington. He succeeds **Capt. Joseph L. Bird,** USN, who has been assigned to the Bureau of Ships in Washington.

P. A. M. Dirac has received the Copley Medal of the Royal Society for his contributions to the relativistic dynamics of a particle in quantum mechanics. The Hughes Medal of the Royal Society was awarded to **P. I. Dee.**

B. D. McDaniel of the physics department, Cornell University, is at the Australian National University, Canberra, on a Fulbright Fellowship.

Special Report Available

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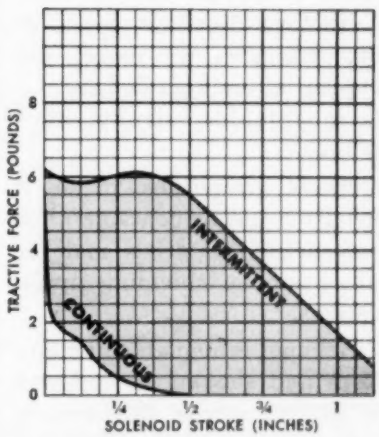

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LETTERS

Hospital Physics in Britain

This letter describing the role of the physicist in British medical establishments was written by Dr. F. W. Spiers, Dept. of Medical Physics, The University of Leeds, England at the request of the editors of NUCLEONICS.

DEAR SIR:

The close association between medicine and physics has long been recognized and is considered of such fundamental importance that a knowledge of general physical principles is regarded as essential to the study of medicine. It is, however, comparatively recently that the physicist has personally become active in various branches of clinical medicine. He entered the hospital via the radiotherapy department, and while his chief sphere of activity remains there, various other departments are recognizing the help he is able to give and are calling on his services.

In Britain, perhaps more than elsewhere, a close working partnership has developed between the radiotherapist and the physicist. Several factors have contributed to this. The radiologist very early realized that he required the help of the physicist if he was to develop the medical use of radiation safely for himself and his patient. Since 1929 the National Radium Commission, in developing a radiotherapy service for the country, did much to concentrate this type of work in the large centers of population and required provision of physical assistance in the main treatment units. The British Institute of Radiology, with its mixed membership of radiologists, physicists and X-ray engineers, has throughout provided a common meeting ground for workers in medical radiology, and an internationally accepted journal to which both doctor and physicist can contribute. Today the hospital physicist is not only accepted by the radiotherapist as an indispensable unit in radiotherapeutic staffing; he also enjoys active support from such bodies as the Faculty of Radiologists, The Royal Society of Medicine, the Medical Research Council and the British Empire Cancer campaign.

It is interesting to consider this university graduate in a "nonmedical" subject, who in so short a period of time has established an almost unique position in the hospital world; to inquire where he works, what he does, how he is trained, about his relationship with the doctor and his service to the patient, his part in research and his association with other scientific workers.

Training and functions. It is essential that the hospital physicist have a first-class knowledge of his own basic subject. In Britain an honors degree is looked for, and some research experience is regarded as an invaluable asset.

Considerable further training is required before the physicist becomes an experienced member of a radiotherapy department. Previous undergraduate study in biology or physiology is of considerable help, but much of his knowledge and experience of clinical medicine and medical terminology must be obtained subsequently at first hand. Most British hospital physicists are indebted to their radiotherapist colleagues for such instruction, although some attempts to provide more formal courses in appropriate medical subjects are now being made by the Hospital Physicists' Association and by some of the larger centers. Although it is too early to suggest that a particular medical syllabus should be laid down as suitable for postgraduate study by the hospital physicist, courses have been held to cover such subjects as radiobiology, radiation chemistry and some aspects of anatomy and physiology. Selection of material must necessarily be made in dealing with subjects as large as these, for, although a general medical knowledge may suffice for many purposes, detailed and much deeper knowledge of a particular medical problem is at times all important. A general indication of the scope of the early courses arranged for hospital physicist by Dr. F. G. Spear is to be found in his recent book (*1*).

An important part of the hospital physicists' training, however, can only be acquired in the clinic itself. On the physical side the hospital physicist must learn to adapt the methods of the laboratory to the needs of the clinic. His apparatus must of necessity be better finished than its counterpart on the laboratory bench. It may need to be capable of sterilization. Above

all it must be reliable even under adverse conditions.

Trained primarily as a scientist, he is likely to find it difficult at first to understand the doctor's apparently unscientific approach to clinical problems wherein the patient's welfare rather than a concise solution is the over-riding consideration. But, without lowering the standard of his own scientific thought, he must school himself always to view the purely physical arguments in perspective with the medical considerations of the sick human being.

Having acquired understanding of the medical and clinical aspects of his work, the physicist gradually becomes an experienced member of a team, able to assess the physical possibilities and limitations of various agents or methods, and to assist in the development of those found suitable for clinical application. His relationship with the doctor eventually becomes that of a partner in procedures that require a combination of special medical and physical knowledge. This is best illustrated by reference to the hospital physicist's daily work.

In many centers the major function of the hospital physicist is to provide the physical control for all forms of radiation therapy. Research is not thereby excluded, but the first duty is to derive and keep up to date the quantitative physical data for the safe and accurate use of radiation sources.

On the one hand this includes the measurement of the output of X-ray tubes and radioactive sources, and the responsibility for radiation monitoring.

On the other hand, in a variety of ways, the physicist may take a direct part in the individual patient's treatment. He may be consulted when the choice of the particular radiation method is under consideration, discussing with the radiotherapist the physical problems of delivering an adequate dose of radiation to the patient's tumor, giving advice and making suggestions in the light of his physical knowledge and clinical experience. In some centers the physicist may then supervise the making of some appliance for giving the treatment, and later will test its efficacy by calculation or by direct radiation measurement. In many cases standardized techniques, worked out by the radiotherapist and physicist together as being applicable to whole groups of patients, can be used, thus avoiding considerable repetition of labor. In others, special difficulties may call for *ad hoc* physical

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investigation to explore the possibilities of a suggested method of treatment before a final decision can be taken.

This participation of the physicist in the planning and evaluation of radiation of treatments is a valuable service to both radiotherapist and patient and ultimately gives the physicist a deep understanding of the clinical and physical problems of cancer treatment with which so much of his work is concerned. Not all the purely computational work need be done by the physicist himself, and it is becoming usual to employ trained technicians to carry out the more routine procedures under the physicist's direction.

The hospital physicist in Britain, already established in X-ray and radium therapy, has naturally played a similar role in the development of the medical use of radioisotopes. In addition to furthering the therapeutic use of radioactive substances other than radium, he has become an essential member of the "radioisotope team" applying radioactive methods to the diagnosis of disease and to the study of body function over a wide field of medicine. He collaborates in devising the methods to be employed, is responsible for the physical measurements (again with the assistance of technicians), and assists in the interpretation of the results.

"Health physics" is not so far regarded as a separate and special activity; only exceptionally is a physicist appointed solely for the purpose of running a hospital protection service. The work is usually carried out as part of the hospital physics service with the aid of a technician.

Considerable advance has been made in the understanding of the mode of action of radiations on living cells. Today there is a vigorous prosecution of research in the fields of radiobiology and radiochemistry. The hospital physicist, with the problems of clinical radiotherapy around him, has so far given only limited attention to this fundamental side of the radiation problem. Much essential information, however, is still lacking on the physical side, and there is great scope for collaboration between the physicist and his colleagues in radiobiology and radiation chemistry.

Organization of hospital physics departments. The organization of hospital physics throughout the country depends to some extent upon the size and character of the institution.

In some London centers hospital physics is directed by the physics

department of the associated medical school. The staff of such a department is large enough to handle teaching of undergraduate and postgraduate medical physics and to allow some members to devote the greater portion of their time to developing the hospital physics services.

There are some institutions of a research character where considerable numbers of physicists are able to work in teams with medical colleagues on intensive studies of special problems. The more general pattern of organization is that common to large radiotherapy centers serving populations of about 1 to 5 million. In these the physics staffs are large enough to provide the radiation physics service and at the same time to devote effort to research and development. Some teaching is also usually done in giving courses in physics for the postgraduate radiological diplomas and the professional diplomas required of radiographers.

The smaller centers frequently have only one physicist on the staff. The cooperation between the radiotherapist and the physicist is then usually very close and effective, but the physicist himself is liable to suffer by the almost unavoidable isolation from other physicists. Attempts are being made in many places to minimize this isolation by associating the small center with the larger center in the region, thereby providing the necessary professional contacts and rendering assistance by the loan of special equipment and workshop services.

The practice of providing an itinerant or visiting physicist for a number of small centers has not been completely satisfactory; it fails chiefly by losing the advantage of having the physicist always present to participate in the day-to-day clinical work, particularly in the treatment of the difficult case.

The distribution of the 188 physicists on the Hospital Physicists' Association register now employed in hospitals, medical schools and biological research departments is shown in the table (p. 82). The average size of staff for all the 71 institutions is nearly three; the number of one-man departments appears rather high, but it includes some new appointments to medical research laboratories and at least 10 sub-centers associated with larger units.

In regions outside London the proportion of physicists to radiotherapists is nearly 1:2. A higher proportion of

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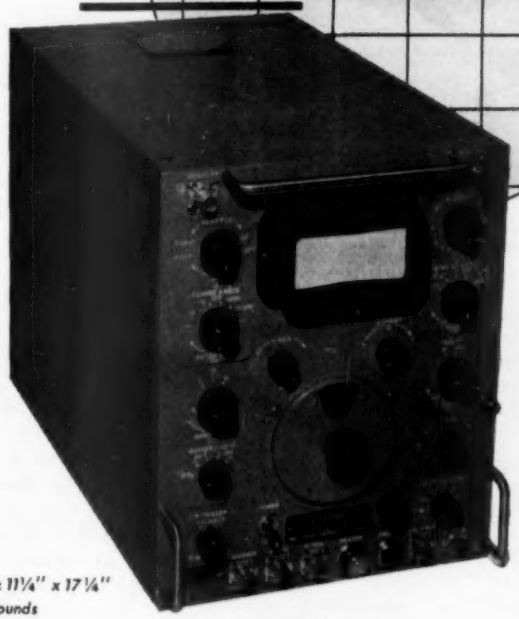

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Distribution of 188 Physicists on 71 British Staffs

Size of staff	*Number of departments*
1	34
2	12
3	6
4	8
5	4
6	2
7	1
8	0
9	1
10	1
11	2

physicists is apparent in the figures for the London area, but this is due to the special conditions in some large research centers where the physicists outnumber the radiotherapists. If we exclude three centers devoted mainly to radiotherapeutic research, the figures for the whole country indicate that one full-time physicist is employed for approximately every two full-time radiotherapists. Earlier statistics compiled by Dr. Margaret Tod (*2*) show that such a "unit" of two radiotherapists and one physicist is needed to carry out the work of a department treating some 500 new cases per year.

It is important that physicists have the assistance of instrument makers and electronics technicians with adequate machine tools and electrical equipment. In the larger centers two instrument makers are considered necessary for every three or four physicists who spend some of their time on research. A similar proportion of electronics technicians is employed in some departments.

Finally, an important development is appearing in some of the larger centers associated with university teaching hospitals, where increasing application of physics is being made to branches of medicine outside radiotherapy (*3*). In one direction, increasing attention is being given to the field of diagnostic radiology, where advances in technology have not only posed new physical problems but also re-introduced some of the older ones in a more acute form (*4*). In other directions, modern electronic techniques have greatly increased our ability to observe transient phenomena and thereby have made possible new studies of body function. The physicist with a knowledge of electronics can find much interest where electrical phenomena are associated with physiological action.

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Problems in the field of electrophysiology, in the study of arterial pressures and in the recording and analysis of heart sounds are among those which may well claim attention. In some centers it has been found advantageous for one or more physicists to devote themselves to the application of electronics, in the widest sense, to both radiological and other medical work.

There is, then, much to be said for one department embracing the wider field of medical physics, even if it must still regard radiation therapy as its major activity. Physicists with a wider variety of skills and interests are thereby held together and mutual advantage comes from the interaction of their work and ideas. If, in addition, there is official connection with the local university, or if, as in some cases, the department becomes a university department, research can be more effectively supported, collaboration with other departments more easily effected, and the important function of training postgraduate physicists undertaken.

The Hospital Physicists' Association maintains a diagrams and data scheme that has assembled scientific data from British centers and made the information readily and cheaply available to any institution requiring it (*5*).

It is clear that the growth of hospital physics in Britain has come about largely as a result of the efforts made to develop the country's radiotherapy services, and that this development itself was greatly facilitated by the existence of relatively compact distributions of population.

The applications of physics to medicine has progressed beyond the stage in which the medical man, in addition to following the increasing complexities of his own specialty, is able also to be his own applied physicist. Collaboration between medical physicist and doctor is to-day essential if the patient is to have the benefit of treatment planned in the light of the most up-to-date medical and physical thought.

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2. M. Tod, "An Inquiry into the Extent to which Cancer Patients in Great Britain Receive Radiotherapy" (John Sherratt & Son, Altringham, England, 1948)
3. F. W. Spiers. The organization of medical radiophysics in Great Britain, *Acta Radiol.* (in press)
4. L. A. W. Kemp, M. H. Jupe. The physicist in the radiodiagnostic department, *Brit. J. Radiol.* **19**, 301 (1946)
5. Hospital physicists' association, formation, *Nature* **152**, 443 (1943); diagrams and data scheme, *Brit. J. Radiol.* **18**, 116 (1945).

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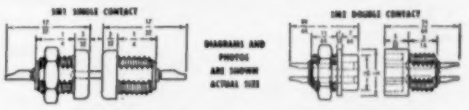

POLARIZATION: Body design of the "SM2" permits engagement in one position only.

RACK & PANEL MOUNTING: Either plug or receptacle may be panel mounted with a ¼-28 cadmium plated brass nut. A melamine hood protects soldered wires.

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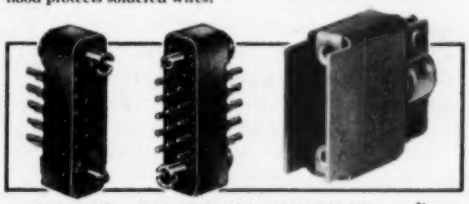
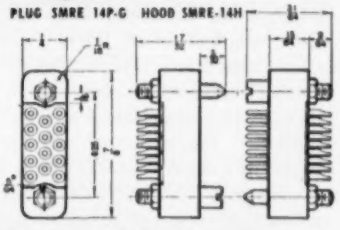


Photo and Drawing of SMRE-14 actual size. Other sizes determined by number of contacts.



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MOUNTING METHOD: *Rivet*, hollow aluminum shank (.094" O.D.) assembled by swaging or spinning; *Tapped Insert*, brass, cadmium plated, clear iridite finish, assembled with No. 4-40 or No. 6-32 standard machine screw; *Threaded Stud*, brass, cadmium plated, clear iridite finish, assembled with No. 4-40 or No. 6-32 nut.

BREAKDOWN VOLTAGE:

Stand-off Body Length	D.C. Voltage
7/32"	7,000 V
3/8"	14,000 V
17/32"	22,000 V
19/32"	24,000 V
Feed-through Creepage Length	
5/32"	4,500 V
9/32"	8,000 V

PHYSICAL AND ELECTRICAL DATA

Recept. Code No.	Plug Code No.	No. of Contacts	Solder Cup *Dia. In.	Weight Oz. Rec.	Weight Oz. Plug	Weight Oz. Hood	D.C. Volts Breakdown Connector Engaged — Sea Level Normal Humidity — Between Contacts	Sea Level Normal Humidity — Contacts to Gnd.	50,000 Ft. Altitude — Between Contacts	50,000 Ft. Altitude — Contacts to Gnd.
SM1S	SM1P	1	.043	.02	.04	.02	....	5400	...	1750
SM2S	SM2P	2	.0225	.02	.02	.02	1600	2600	800	1100
SMRE 7S-G	SMRE 7P-G	7	.0225	.1	.1	.1	2450	2050	950	675
SMRE 14S-G	SMRE 14P-G	14	.0225	.1	.2	.1	2450	2050	950	675
SMRE 20S-G	SMRE 20P-G	20	.0225	.2	.2	.2	2450	2050	950	675
SMRE 26S-G	SMRE 26P-G	26	.0225	.2	.2	.2	2450	2050	950	675
SMRE 29S-G	SMRE 29P-G	29	.0225	.2	.2	.2	2450	2050	950	675
SMRE 34S-G	SMRE 34P-G	34	.0225	.2	.2	.2	2450	2050	950	675

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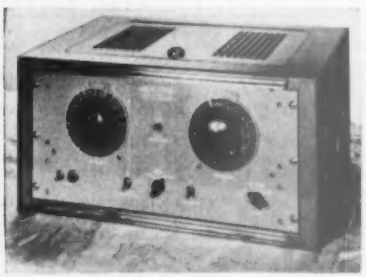
Radioactive Products, Inc., 443 W. Congress, Detroit 26, Mich. This set of punched cards, utilizing the McBee Keysort system (NU, Feb. '53, p. 68), classifies the properties of almost 1,200 nuclides. Radioisotopes can be selected by half-life (42 subclasses); alpha (9 subclasses), beta (19 subclasses), and gamma energy (19 subclasses); and availability. The system is supplied with sorting trays and probes. Revisions will be offered, but are not expected to be extensive. (P-2)



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Radiation Instrument Development Laboratory, 2337 W. 67th St., Chicago 36, Ill. The model 115 single-channel pulse-height analyzer handles pulses of 0–100 volt amplitude. The unit contains linear amplifier, coincidence and anticoincidence circuits, and pulse generator. Channel width is adjustable from 0 to 10 volts; switch allows 0–5 or 0–10 volt ranges. Channel width and channel level are calibrated to read directly in volts. Amplifier gain is 1,000; amplifier attenuator settings are 1, ½, ¼, ⅛, and 1/16. Pulse generator is self contained for checking calibration. The unit may be used as an integral or differential pulse-height analyzer. (P-3)

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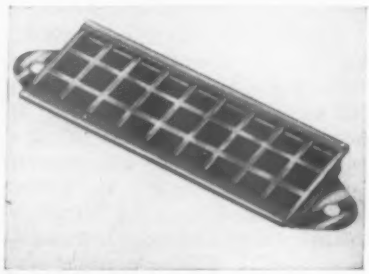

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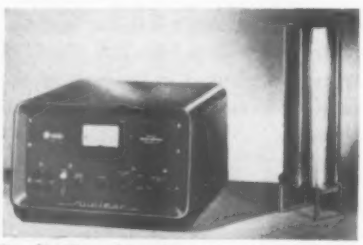

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Nuclear Instrument and Chemical Corp., 229 W. Erie St., Chicago 10, Ill. The model 1310 area monitor, developed at ORNL, consists of an ion chamber and a main chassis. It is used for measurement of relatively

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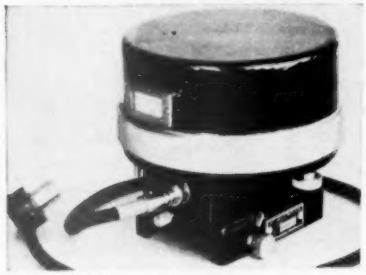

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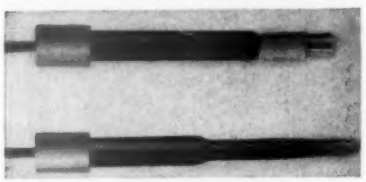

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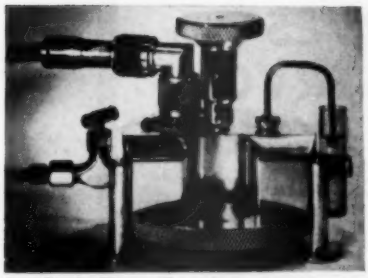

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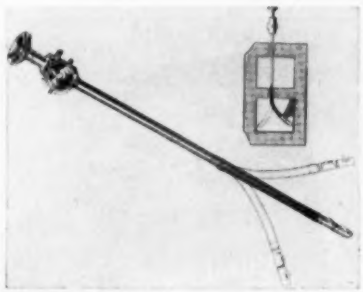
Atomic Center for Instruments & Equipment, Inc., 489 Fifth Ave., New York 17, N. Y. The model 21 preflush flow counter is a 3-position windowless counter for operation in the Geiger and proportional regions. The sample platform contains recesses for loading, preflushing, and counting. In the Geiger region with helium-butane mixture, the plateau is 200 volts, slope is <4%, and starting potential is 1,250 volts. For proportional counting with methane gas, the plateau is 300 volts, slope <1%, and starting potential is 3,000 volts. Background is 18 cpm. Well size is $1\frac{9}{32} \times \frac{3}{8}$ in. (P-11)



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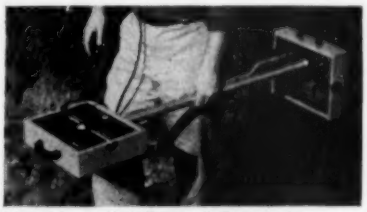
Westinghouse Electric Corp., 401 Liberty Ave., Pittsburgh 30, Pa. Types WX-3347 and WX-4813 transistors are available in sample lots for development use in amplifier, oscillator, and switching circuits. As a grounded-base amplifier under small signal conditions, the point-contact-type WX-3347 operating characteristics are: collector current, 2–3 ma; power gain, 18 db; cutoff frequency, 2 Mc. As an amplifier with grounded emitter and base input, P-N-P junction-type WX-4813 characteristics are: collector current, 1–2 ma; power gain, 30 db. (P-13)



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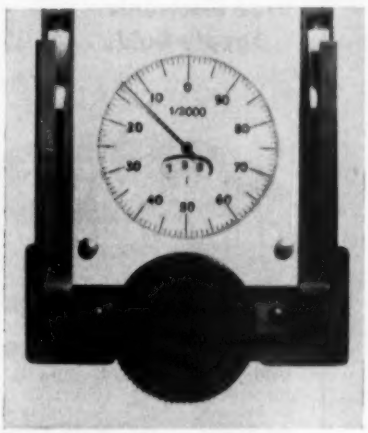

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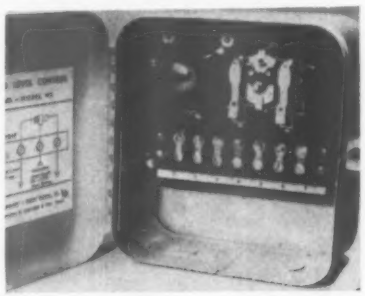

Liquid-Level Control

Haledy Electronics Co., 57 William St., New York 5, N. Y. This electronic liquid-level control utilizes an all-cold-cathode-tube design. Liquid contact is made by stainless-steel rods. Accuracy is independent of pressure and temperature. The Haledy TT-1 triode used has an amplification factor of $2\frac{1}{2} \times 10^6$. Current across the electrodes is <2 μa. Electrodes can be up to 10 ft; suspended probes can be used for greater lengths. (P-16)

Nonindicating Meter-Relay

Assembly Products, Inc., Box 191, Chagrin Falls, Ohio. The nonindicating meter-relay is a moving-coil relay with self-locking contacts. Accuracy is better than 1% of specified current or voltage for most values. Operating ranges are 0.2 μa to 50 amperes and 0.05 mv to 500 volts; higher values can be handled with external resistors. Internal or external rectifiers can be

supplied for a-c applications. Normal contacts have ratings up to 100 ma, d-c, for "make" only; heavy-duty contacts are available. (P-17)

Relay Controls

Para Laboratory Supply Co., 221 N. Hermitage Ave., Trenton, N. J. These relay circuits are designed for low-sensing-current operation of sensing elements used in temperature, liquid-level, and flame control and in automatic gaging. With the relays, sensing elements do not carry more than 0.1 μa through a maximum resistance of 100 megohms; modified circuits are available where the maximum current may be 0.01 or 0.001 μa through a maximum resistance of 500 megohms. Various models are available with 2 or 3 load receptacles and load capacities of 15 or 30 amperes. Models are provided with or without 3-position switches for off, normally open, and normally closed operation. (P-18)

R-F Power Supplies

Neutronic Associates, 83-56 Vietor Ave., Elmhurst 73, N. Y. These power supplies may be reversed in polarity by a lever on the front panel. Models R-22C and R-22M, unregulated, have ranges of 3-30 kv with 2 ma at 18 kv and 3 ma at 20 kv, respectively. Models R-22CR and R-22MR, regulated to 0.5%, have a 5-30 kv range with same current capabilities. (P-19)

Transistors

RCA Victor Div., Radio Corporation of America, Camden, N. J. Two point-contact and two junction-type transistors are available. Point-contact types are RCA-2N32 for large-signal applications, such as pulse or switching service, computers, counters, and on-off controls, and RCA-2N33 for use as an oscillator at frequencies up

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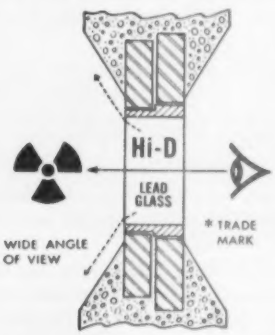

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to 50 Mc. RCA-2N34, P-N-P, and RCA-2N235, N-P-N, junction types are designed for low-power audio-frequency applications. (P-20)

Reversible Balancing Motor

Industrial Div., Minneapolis-Honeywell Regulator Co., Wayne & Windrim Aves., Philadelphia 44, Pa. This reversible two-phase balancing motor, rated at 333 rpm, no-load, can be applied to recording instruments, remote-control devices, and other apparatus requiring positive positioning by a motor operated from an electronic amplifier. It provides sufficient torque to drive a recorder pen carriage full scale in 1.8 sec. The motor is self lubricated; an air vent relieves the oil seal from pressure. It is designed for use at 20–175° F. (P-21)

High-Voltage-Breakdown Tester

Industrial Instruments, Inc., 89 Commerce Rd., Cedar Grove, N. J. The model P-7-20 high-voltage-breakdown tester supplies 0–20,000 volts, a-c or d-c, at low current drain. Maximum d-c current is 15 ma. Cutoff control is provided so that power is turned off when load current exceeds any preset value from 5 to 20 ma. Load current, d-c voltage and peak a-c voltage are read directly. (P-22)

Magnetic Tape Recorder

Raytheon Mfg. Co., Waltham 54, Mass. This computer magnetic tape mechanism, for recording and storage of digital data, may be operated automatically by pulsed input signals or controlled manually by a front-panel switch. The mechanism moves from stop to start or vice versa in <5 msec. The magnetic head has six narrow-gap channels with high-frequency response. Power supply is built in. Slack and tension control of the tape is automatic. Two 1,000-ft reels are mounted on coaxially-driven shafts. (P-23)

LITERATURE AVAILABLE

Electronic instruments. Catalog gives specifications on line of power supplies, oscilloscopes, rheostats and micrometers. *Beta Electric Corp., 333 E. 103rd St., New York 29, N. Y.* (L-1)

Photo-recording manual. "Techniques of Photo-Recording from Cathode-Ray Tubes" reviews problems and associated solutions encountered with

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cathode-ray patterns. *Allen B. Du Mont Laboratories, Inc., 760 Bloomfield Ave., Clifton, N. J.* (L-2)

Weighing equipment. Bulletin 15C illustrates and describes line of scales, balances, weights, and accessories. *Central Scientific Co., 1700 Irving Park Rd., Chicago 13, Ill.* (L-3)

Spectrophotometry. "What Every Executive Should Know about Spectrophotometry" tells what a spectrophotometer is, how it works, and what to look for in selecting an instrument. *Beckman Instruments, Inc., S. Pasadena, Calif.* (L-4)

Measuring equipment. Catalog GEC-1016A contains information on more than 115 testing and measuring devices for laboratory and production-line use. *General Electric Co., Schenectady 5, N. Y.* (L-5)

Glass-bonded mica. "From One Machinist to Another" tells how to machine glass-bonded mica. *Mycalex Corp. of America, Clifton Blvd., Clifton, N. J.* (L-6)

"Dag" dispersions. Pamphlet lists all current dispersions, including colloidal graphite, molybdenum disulfide, vermiculit, and zinc oxide. *Acheson Colloids Co., Port Huron, Mich.* (L-7)

Remote-control equipment. Pamphlet describes "Multi-Gate" remote supervisory control system for organizations that have widespread facilities. *Hammarlund Mfg. Co., Inc., 460 W. 34th St. New York 1, N. Y.* (L-8)

Electroplating components. Folder describes "Cantavone" process for volume electroplating precision components. *American Electro Products, Inc., 1358 Thomaston Ave., Waterbury, Conn.* (L-9)

Induction-motor starters. Bulletin 14B7733 tells how to select the right starter for squirrel-cage induction motors rated up to 600 hp at 600 volts or less. *Allis-Chalmers Mfg. Co., 1316 S. 70th St., Milwaukee, Wis.* (L-10)

Laboratory electronic equipment. Catalog gives specifications on attenuation networks, resistor units, volume-level indicators, transmission-measuring sets, amplifiers, frequency meters, etc. *Daven Co., 191 Central Ave., Newark 4, N. J.* (L-11)

Aliphatic amides. "Armids" describes chemical properties and applications of these high molecular weight fatty acid derivatives. *Armour and*

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Co., Armour Chemical Div., 1355 W. 31st St., Chicago 9, Ill. (L-12)

Ceramic properties. Chart 531 lists mechanical and electrical properties of AlSiMag ceramics. *American Lava Corp., Chattanooga 5, Tenn.* (L-13)

Plastic pipe. Folder gives specifications of corrosion-resistant plastic pipe. *Anesite Co., 3575 Touhy Ave., Chicago 45, Ill.* (L-14)

Filters. Sheet describes cylinder-type disk filter and down-flow filter; double closure permits use of various filter media. *Ertel Engineering Corp., Kingston, N. Y.* (L-15)

Liquid-level control. Folder LL4-453 describes single-thyratron electronic liquid-level control operating without radio frequency from a single-capacitive-type probe. *Thermo Instruments Co., 1166 El Camino Real, Belmont, Calif.* (L-16)

High-purity copper. Sheet gives data on Cuprovac-E, gas-free high-purity copper with properties suited for vacuum-tube manufacture. *National Research Corp., 70 Memorial Dr., Cambridge, Mass.* (L-17)

Wire-wound resistors. Bulletin C-1 gives data on tubular and flat resistors. *International Resistance Co., 401 N. Broad St., Philadelphia 8, Pa.* (L-18)

Electroplated gold. Pamphlet gives information on electroplating gold together with other properties of gold. *Technic, Inc., 39 Snow St., Providence, R. I.* (L-19)

Mixed-bed demineralizers. Bulletin 126 explains how a mixed-bed demineralizer operates in producing high-purity water by ion-exchange method. *Barnstead Still & Sterilizer Co., 222 Lanesville Terrace, Forest Hills, Boston 31, Mass.* (L-20)

Vacuum furnace. Catalog 770 discusses processes now being carried out in the 3 types of vacuum furnaces being built. *F. J. Stokes Machine Co., Vacuum Equipment Div., 5500 Tabor Rd., Philadelphia 20, Pa.* (L-21)

Temperature controls. Bulletin 101 describes controls for high-temperature safety alarms and cut-outs up to 1,800° F. *Burling Instrument Co., 5 Vose Ave., South Orange, N. J.* (L-22)

High-vacuum pumps. Catalog 750 includes tables of formulas, constants, and conversions used in vacuum processing; solutions to problems of pump

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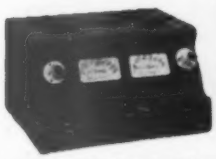

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selection; and information on oil purification and other pump maintenance. *F. J. Stokes Machine Co., 5500 Tabor Rd., Philadelphia 20, Pa.* (L-23)

Galvanometer selection. Bulletin is guide to selection of galvanometers and recording oscillographs for use with pressure and temperature pickups. *Trans-Sonics, Inc., Bedford Airport, Bedford, Mass.* (L-24)

Furnace and oven control. Catalog P1255 describes potentiometer and millivoltmeter type pyrometer controllers and indicators. *The Bristol Co., Waterbury 20, Conn.* (L-25)

Recording sulfur concentration. Bulletin CEC 1810B describes Titrilog cabinet and portable models for continuously recording concentration of sulfur compounds in gas streams or atmosphere. *Consolidated Engineering Corp., 300 N. Sierra Madre Villa, Pasadena 15, Calif.* (L-26)

Strain-indicator. Bulletin 4103 describes portable strain indicator. *Baldwin-Lima-Hamilton Corp., Philadelphia 42, Pa.* (L-27)

Shock machine. Bulletin 533 presents data on machine for calibrated shock tests up to 77g peak acceleration. *The Barry Corp., 879 Pleasant St., Watertown 72, Mass.* (L-28)

Corrosion-resistant chemical equipment. Bulletin BMA-5 3-53 5M describes Chemplas phenolic resin piping, towers, pumps, etc., for high-temperature corrosion resistance. *General Ceramics and Steatite Corp., Chemical Equipment Div., Keasbey, N. J.* (L-29)

INDUSTRY NOTES

▶ **Atomic Instrument Co.,** 84 Massachusetts Ave., Cambridge, Mass., has appointed Rudolph Grave, A/B, Stockholm, Sweden, to handle sales in Sweden and Norway, and Eastern Electric & Engineering Co., Bombay, to handle sales in India.

▶ **Consolidated Engineering Corp.,** Analytical Service Dept., 300 N. Sierra Madre Villa, Pasadena 8, Calif., offers a custom leak-detection service for tests of either vacuum or pressure prototypes.

▶ **Radiation Instrument Development Laboratory,** 2337 W. 67th St., Chicago 36, Ill., has expanded its glass-working facilities for the manufacture of special items, including liquid-scintillation-phosphor containers, counter shells, and Marinelli beakers.

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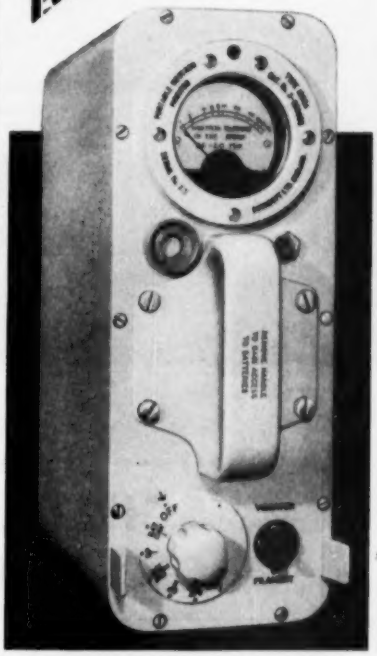

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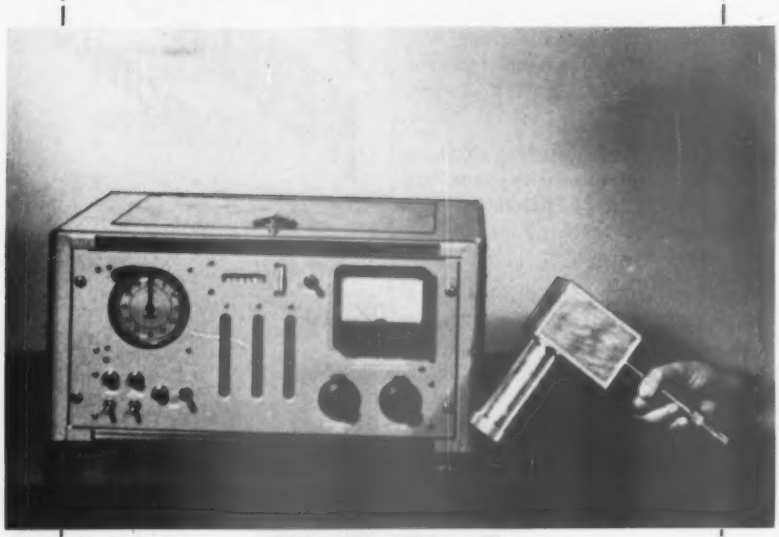

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INDEX TO ADVERTISERS

October, 1953

Alden Products Co. 74
Allied Radio Corp. 86
American Phenolic Corp. 62
Atomic Instrument Co. . . . . 3rd Cover
Bar-Ray Products Inc. 57
Berkeley Division Beckman Instruments, Inc. 79
Bersworth Chemical Co. 88
Burndept Ltd. 91
Cannon Electric Company 77
Cambridge Thermonic Corporation 4
Carboloy, Department of General Electric Company 78
Century Geophysical Corp. 67
Chicago Transformer, Div. of Essex Wire Corp. 89
Consolidated Vacuum Corporation . . . . . 2nd Cover
Detectolab, Inc. 80
Ekco Electronics Ltd. 71
Electrical & Physical Instrument Corp. . . . . 82 & 91
Ford Instrument Company 86
General Electric Co. 6
General Mills, Inc. Mechanical Div. 82
Hanovia Chemical & Mfg. Co. . . . 76
Harshaw Scientific, Div. of the Harshaw Chemical Co. 65
International Resistance Co. 3
Kewaunee Mfg. Co. 87
Kinney Manufacturing Co. 5
Kollmorgen Optical Corp. 90
McGraw-Hill Book Co., Technical Writing Service 90
Michigan Chemical Corporation . . 80
Minneapolis-Honeywell Regulator Co., Industrial Division 61
Nuclear Instrument & Chemical Corp. . . . . 4th Cover
Penberthy Instrument Co. 88
Precise Measurements Co. 91
Radiation Instrument Development Laboratory 92
Rockbar Corporation 88
Special Screw Products Co. 64
Star Stainless Screw Co. 82
Technical Associates 75
Technical Service Corporation . . . 80
Titeflex, Inc. 63
Victoreen Instrument Co. 69
Waterman Products Co., Inc. 81
Where to Buy 91
Winchester Electronics Inc. 83

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● Model 1030A has the same features as Model 1050A, except that it has 0.25 volt input sensitivity only.

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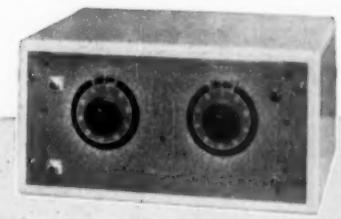

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